

## TRANSLATION CERTIFICATE



I, Tamotsu Ohtani, hereby certify that:

1. I am a citizen of Japan residing at Bridgestone Toranomon Building 6F, 25-2, Toranomon 3-chome, Minato-ku, Tokyo Japan.
2. To the best of my ability, I translated the Japanese language specification, claims and abstract included in

United States Patent Application No. 10/617,397

into English and the attached document is a true and accurate English language translation thereof.

3. I declare further that all statements made herein on personal knowledge are true and that all statements made on information and belief are believed to be true; and further that these statements were made with the knowledge that willful false statements and the like so made are punishable by fine or imprisonment, or both, under Section 1001 of Title 18 of the United States Code and that such willful false statements may jeopardize the validity of the application or any patent issuing thereon.

Date: October 20, 2003

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## DESCRIPTION

### ORGANIC ELECTROLUMINESCENCE DEVICE AND ORGANIC LIGHT EMITTING MEDIUM

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#### TECHNICAL FIELD

The present invention relates to an organic electroluminescence (“electroluminescence” will be referred to as “EL”, hereinafter) device and an organic light emitting medium, and more particularly, to an organic EL device which exhibits excellent purity of color, has excellent heat resistance and a long life and efficiently emits bluish to yellowish light and an organic light emitting medium which is advantageously used for the organic EL device.

#### 15 BACKGROUND ART

An organic EL is a spontaneous light emitting device which utilizes the principle that a fluorescent substance emits light by energy of recombination of holes injected from an anode and electrons injected from a cathode when an electric field is applied.

20 Since an organic EL device of the laminate type driven under a low electric voltage was reported by C. W. Tang of Eastman Kodak Company (C. W. Tang and S. A. Vanslyke, Applied Physics Letters, Volume 51, Pages 913, 1987), many studies have been conducted on organic EL devices using organic materials as the constituting materials.

25 Tang et al used a laminate structure using tris(8-hydroxy-quinolinato)aluminum for the light emitting layer and a triphenyldiamine

derivative for the hole transporting layer. Advantages of the laminate structure are that the efficiency of hole injection into the light emitting layer can be increased, that the efficiency of forming excited particles which are formed by blocking and recombining electrons injected from the 5 cathode can be increased and that excited particles formed within the light emitting layer can be enclosed. As the structure of the organic EL device, a two-layered structure having a hole transporting (injecting) layer and an electron transporting and light emitting layer and a three-layered structure having a hole transporting (injecting) layer, a light emitting 10 layer and an electron transporting (injecting) layer are well known. To increase the efficiency of recombination of injected holes and electrons in the devices of the laminate type, the structure of the device and the process for forming the device have been studied.

As the light emitting material, chelate complexes such as tris(8-15 quinolinato)aluminum, coumarine derivatives, tetraphenyl- butadiene derivatives, bisstyrylarylene derivatives and oxadiazole derivatives are known. It is reported that light in the visible region ranging from blue light to red light can be obtained by using these light emitting materials and development of a device exhibiting color images is expected (For 20 example, Japanese Patent Application Laid-Open Nos. Heisei 8(1996)-239655, Heisei 7(1995)-138561 and Heisei 3(1991)-200289).

Devices using bisanthracene derivatives as the hole transporting material or the light emitting material are disclosed in the United States Patent No. 3008897 and Japanese Patent Application Laid-Open No. 25 Heisei 8(1996)-12600. Although bisanthracene derivatives can be used as the material emitting blue light, the efficiency of light emission and the life

are insufficient for practical applications. In Japanese Patent Application Laid-Open No. 2001-207167, a device using an aminoanthracene derivative as the material emitting green light is disclosed. However, the organic EL device using this material cannot be used for the practical 5 applications since the device has poor heat resistance due to the low glass transition temperature of the material and light emission of a long life and a high efficiency cannot be obtained. Other organic devices of a long life and excellent properties are recently reported. However, the life and the properties are not always sufficient. Therefore, the development of an 10 organic EL device exhibiting more excellent properties have been strongly desired.

#### DISCLOSURE OF THE INVENTION

Under the above circumstances, the present invention has an object 15 of providing an organic EL device which exhibits a high purity of color, has excellent heat resistance and a long life and efficiently emits bluish to yellowish light and an organic light emitting medium which can be advantageously used for the organic EL device.

As the result of extensive studies by the present inventors to achieve 20 the above object, it was found that, when an organic light emitting medium comprised a combination of a specific arylamine compound and at least one compound selected from specific anthracene derivatives, spirofluorene derivatives, compounds having condensed rings and metal complex compounds and an organic electroluminescence device had a layer of the 25 medium disposed between a pair of electrodes, the organic EL device exhibited a high purity of color, had excellent heat resistance and a long

life and efficiently emitted bluish to yellowish light. The present invention has been completed based on this knowledge.

The present invention provides an electroluminescence device comprising a pair of electrodes and a layer of an organic light emitting medium disposed between the pair of electrodes, wherein the layer of an organic light emitting medium comprises:

(A) at least one compound selected from substituted and unsubstituted arylamines having 10 to 100 carbon atoms, and

(B) at least one compound selected from:

10 anthracene derivatives represented by following general formula (I):



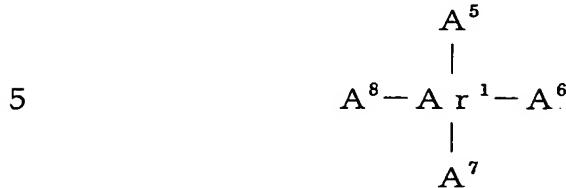
wherein  $A^1$  and  $A^2$  each independently represent a substituted or unsubstituted monophenylanthryl group or a substituted or unsubstituted 15 diphenylanthryl group and may represent a same group or different groups, and  $L$  represents a single bond or a divalent bonding group,

anthracene derivatives represented by following general formula (II):



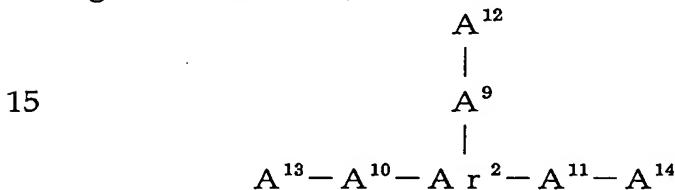
20 wherein  $A_n$  represents a substituted or unsubstituted divalent anthracene residue group,  $A^3$  and  $A^4$  each independently represent a substituted or unsubstituted aryl group having 6 to 40 carbon atoms, at least one of  $A^3$  and  $A^4$  represents a substituted or unsubstituted monovalent condensed aromatic ring group or a substituted or unsubstituted aryl group having 10 25 or more carbon atoms, and  $A^3$  and  $A^4$  may represent a same group or different groups,

spirofluorene derivatives represented by following general formula (III):



wherein  $A_{r^1}$  represents a substituted or unsubstituted spirofluorene residue group,  $A^5$  to  $A^8$  each independently represent a substituted or 10 unsubstituted aryl group having 6 to 40 carbon atoms,

compounds having condensed rings represented by following general formula (IV):



wherein  $A_{r^2}$  represents a substituted or unsubstituted aromatic ring group having 6 to 40 carbon atoms,  $A^9$  to  $A^{11}$  each independently represent a 20 substituted or unsubstituted arylene group having 6 to 40 carbon atoms,  $A^{12}$  to  $A^{14}$  each independently represent hydrogen atom, an alkyl group having 1 to 6 carbon atoms, a cycloalkyl group having 3 to 6 carbon atoms, an alkoxy group having 1 to 6 carbon atoms, an aryloxy group having 5 to 18 carbon atoms, an aralkyloxy group having 7 to 18 carbon atoms, an 25 arylamino group having 5 to 16 carbon atoms, nitro group, cyano group, an ester group having 1 to 6 carbon atoms or a halogen atom, and at least one of  $A^9$  to  $A^{14}$  represents a group having condensed aromatic rings, and

metal complex compounds. The present invention also provides an electroluminescence device described above, wherein 30 component (B) is at least one compound selected from the anthracene

derivatives represented by general formulae (I) and (II) shown above.

The present invention provides an organic light emitting medium which comprises (A) at least one compound selected from substituted and unsubstituted arylamines having 10 to 100 carbon atoms and (B) at least one compound selected from anthracene derivatives represented by general formula (I) shown above, anthracene derivatives represented by following general formula (II) shown above, spirofluorene derivatives represented by general formula (III) shown above, compounds having condensed rings represented by general formula (IV) shown above and metal complex compounds described above. The present invention also provides an organic light emitting medium described above, wherein component (B) is at least one compound selected from the anthracene derivatives represented by general formulae (I) and (II) shown above.

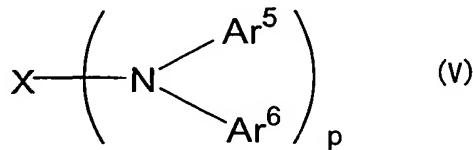
15 THE MOST PREFERRED EMBODIMENT TO CARRY OUT THE INVENTION

The organic EL device of the present invention is a device having a structure comprising a pair of electrodes and a layer of an organic light emitting medium disposed between the pair of electrodes.

20 In the present invention, an organic light emitting medium comprising a combination of (A) at least one compound selected from substituted and unsubstituted arylamine compounds having 10 to 100 carbon atoms and (B) at least one compound selected from the anthracene derivatives represented by the above general formula (I), the anthracene derivatives represented by the above general formula (II), the spirofluorene derivatives represented by the above general formula (III),

the compounds having condensed rings represented by the above general formula (IV) and the above metal complex compounds, is used in the layer of an organic light emitting medium.

Examples of the arylamine compound of component (A) include 5 arylamine compounds represented by the following general formula (V):



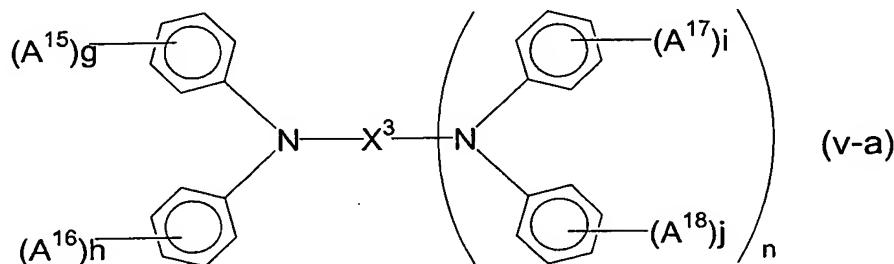
wherein  $\text{X}^3$  represents a substituted or unsubstituted condensed aromatic ring group having 10 to 40 nuclear carbon atoms,  $\text{Ar}^5$  and  $\text{Ar}^6$  each independently represent a substituted or unsubstituted monovalent aromatic group having 6 to 40 carbon atoms, and  $p$  represents an integer of 10 1 to 4.

In general formula (V), examples of the group represented by  $\text{X}^3$  include residue groups derived from naphthalene, phenanthrene, fluoranthene, anthracene, pyrene, perylene, coronene, chrysene, picene, diphenylanthracene, fluorene, triphenylene, rubicene, benzoanthracene, 15 phenylanthracene, bisanthracene, dianthracenylbenzene and dibenzoanthracene. Among these groups, residue groups derived from chrysene, pyrene and anthracene are preferable.

Examples of the monovalent aromatic group having 6 to 40 carbon atoms which is represented by  $\text{Ar}^5$  and  $\text{Ar}^6$  include phenyl group, naphthyl group, anthranyl group, phenanthryl group, pyrenyl group, coronyl group, biphenyl group, terphenyl group, fluorenyl group, furanyl group, thieryl group, benzothienyl group, indolyl group and carbazolyl group. Among these groups, phenyl group, naphthyl group, pyrenyl group and biphenyl

group are preferable.

As the arylamine compound represented by general formula (V), arylamine compounds represented by the following general formula (V-a):



wherein X<sup>3</sup> is as defined in general formula (V), are preferable.

5 In the above general formula (V-a), Ar<sup>15</sup> to Ar<sup>18</sup> each independently  
represent hydrogen atom, a substituted or unsubstituted alkyl group  
having 1 to 50 carbon atoms and preferably 1 to 20 carbon atoms, a  
substituted or unsubstituted aryl group having 5 to 50 carbon atoms and  
preferably 5 to 20 carbon atoms, a substituted or unsubstituted aralkyl  
10 group having 7 to 50 carbon atoms and preferably 7 to 40 carbon atoms, a  
substituted or unsubstituted cycloalkyl group having 3 to 50 carbon atoms  
and preferably 5 to 12 carbon atoms, a substituted or unsubstituted  
alkoxyl group having 1 to 50 carbon atoms and preferably 1 to 6 carbon  
atoms, a substituted or unsubstituted aryloxyl group having 5 to 50 carbon  
15 atoms and preferably 5 to 18 carbon atoms, a substituted or unsubstituted  
arylarnino group having 5 to 50 carbon atoms and preferably 5 to 18 carbon  
atoms or a substituted or unsubstituted alkylarnino group having 1 to 20  
carbon atoms and preferably 1 to 6 carbon atoms.

Examples of the substituted or unsubstituted alkyl group  
20 represented by Ar<sup>15</sup> to Ar<sup>18</sup> include methyl group, ethyl group, propyl  
group, isopropyl group, butyl group, sec-butyl group, tert-butyl group,

pentyl group, hexyl group, heptyl group, octyl group, stearyl group, 2-phenylisopropyl group, trichloromethyl group, trifluoromethyl group, benzyl group,  $\alpha$ -phenoxybenzyl group,  $\alpha,\alpha$ -dimethylbenzyl group,  $\alpha,\alpha$ -methylphenylbenzyl group,  $\alpha,\alpha$ -ditrifluoromethylbenzyl group, 5-triphenylmethyl group and  $\alpha$ -benzyloxybenzyl group.

Examples of the substituted or unsubstituted aryl group represented by Ar<sup>15</sup> to Ar<sup>18</sup> include phenyl group, 2-methylphenyl group, 3-methylphenyl group, 4-methylphenyl group, 4-ethylphenyl group, biphenyl group, 4-methylbiphenyl group, 4-ethylbiphenyl group, 4-cyclohexylbiphenyl group, terphenyl group, 3,5-dichlorophenyl group, naphthyl group, 5-methylnaphthyl group, anthryl group and pyrenyl group.

Examples of the substituted or unsubstituted aralkyl group represented by Ar<sup>15</sup> to Ar<sup>18</sup> include benzyl group, 1-phenylethyl group, 2-phenylethyl group, 1-phenylisopropyl group, 2-phenylisopropyl group, phenyl-t-butyl group,  $\alpha$ -naphthylmethyl group, 1- $\alpha$ -naphthylethyl group, 2- $\alpha$ -naphthylethyl group, 1- $\alpha$ -naphthylisopropyl group, 2- $\alpha$ -naphthylisopropyl group,  $\beta$ -naphthylmethyl group, 1- $\beta$ -naphthylethyl group, 2- $\beta$ -naphthylethyl group, 1- $\beta$ -naphthylisopropyl group, 2- $\beta$ -naphthylisopropyl group, 1-pyrrolylmethyl group, 2-(1-pyrrolyl)ethyl group, p-methylbenzyl group, m-methylbenzyl group, o-methylbenzyl group, p-chlorobenzyl group, m-chlorobenzyl group, o-chlorobenzyl group, p-bromobenzyl group, m-bromobenzyl group, o-bromobenzyl group, p-iodobenzyl group, m-iodobenzyl group, o-iodobenzyl group, p-hydroxybenzyl group, m-hydroxybenzyl group, o-hydroxybenzyl group, p-aminobenzyl group, m-aminobenzyl group, o-aminobenzyl group, p-

nitrobenzyl group, m-nitrobenzyl group, o-nitrobenzyl group, p-cyanobenzyl group, m-cyanobenzyl group, o-cyanobenzyl group, 1-hydroxy-2-phenylisopropyl group and 1-chloro-2-phenylisopropyl group.

Examples of the substituted or unsubstituted cycloalkyl group represented by Ar<sup>15</sup> to Ar<sup>18</sup> include cyclopropyl group, cyclobutyl group, cyclopentyl group and cyclohexyl group.

Examples of the substituted or unsubstituted alkoxy group represented by Ar<sup>15</sup> to Ar<sup>18</sup> include methoxyl group, ethoxyl group, propoxyl group, isopropoxyl group, butoxyl group, isobutoxyl group, sec-10 butoxyl group, tert-butoxyl group, various types of pentyloxy groups and various types of hexyloxy groups.

Examples of the substituted or unsubstituted aryloxy group represented by Ar<sup>15</sup> to Ar<sup>18</sup> include phenoxy group, tolyloxy group and naphthoxy group.

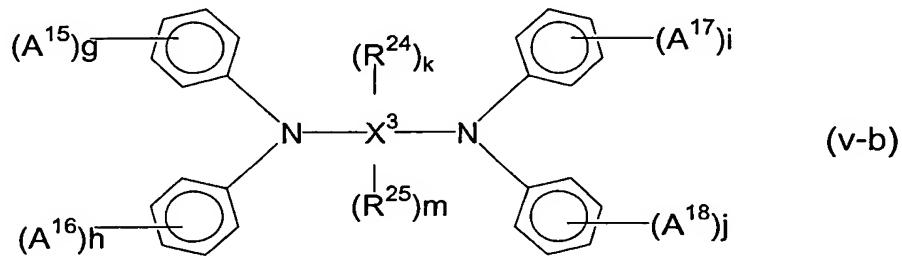
15 Examples of the substituted or unsubstituted arylamino group represented by Ar<sup>15</sup> to Ar<sup>18</sup> include diphenylamino group, ditolylamino group, dinaphthylamino group and naphthylphenylamino group.

Examples of the substituted or unsubstituted alkylamino group represented by Ar<sup>15</sup> to Ar<sup>18</sup> include dimethylamino group, diethylamino group and dihexylamino group.

In general formula (V-a), g, h, i and j each represent an integer of 0 to 5 and preferably an integer of 0 to 2. Atoms and groups represented by a plurality of Ar<sup>15</sup> to Ar<sup>18</sup> may be the same with or different from each other and may be bonded to each other to form a saturated or unsaturated 25 ring when g, h, i and j each represent an integer of 2 or greater. n represents an integer of 0 to 3. At least one of Ar<sup>15</sup> to Ar<sup>18</sup> represents a

substituted or unsubstituted secondary or tertiary alkyl group having 3 to 10 carbon atoms. Examples of the secondary or tertiary alkyl group include the secondary and tertiary alkyl groups among the groups described as the examples of the alkyl group represented by Ar<sup>15</sup> to Ar<sup>18</sup>.

5 As the arylamine compound represented by general formula (V), arylamine compounds represented by the following general formula (V-b):



10 wherein X<sup>3</sup>, Ar<sup>15</sup> to Ar<sup>18</sup>, g, h, i and j are as defined in general formula (V-a), and atoms and groups represented by a plurality of Ar<sup>15</sup> to Ar<sup>18</sup> may be the same with or different from each other and may be bonded to each other to form a saturated or unsaturated ring when g, h, i and j each represent an integer of 2 or greater, are also preferable.

15 In general formula (V-b), R<sup>24</sup> and R<sup>25</sup> each independently represent hydrogen atom, a substituted or unsubstituted alkyl group having 1 to 10 carbon atoms and preferably 1 to 6 carbon atoms, a substituted or unsubstituted aryl group having 6 to 20 carbon atoms and preferably 6 to 14 carbon atoms, a substituted or unsubstituted aralkyl group having 7 to 50 carbon atoms and preferably 7 to 40 carbon atoms, a substituted or unsubstituted alkoxy group having 1 to 50 carbon atoms and preferably 1 to 6 carbon atoms or a substituted or unsubstituted aryloxy group having 20 5 to 50 carbon atoms and preferably 5 to 18 carbon atoms.

Examples of the groups represented by R<sup>24</sup> and R<sup>25</sup> include the

groups in which the number of carbon atoms is within the above range among the groups described as the examples of the groups represented by Ar<sup>15</sup> to Ar<sup>18</sup>.

At least one of R<sup>24</sup> and R<sup>25</sup> represents a substituted or 5 unsubstituted secondary or tertiary alkyl group having 3 to 10 carbon atoms. Examples of the secondary or tertiary alkyl group include the secondary and tertiary alkyl groups among the groups described as the examples of the alkyl group represented by Ar<sup>15</sup> to Ar<sup>18</sup>.

In general formula (V-b), k and m each represent an integer of 0 to 10 2.

In the present invention, the arylamine compound of component (A) may be used singly or in combination of two or more.

Examples of the substituent in the compounds represented by general formulae (V), (V-a) and (V-b) include alkyl groups having 1 to 6 15 carbon atoms, cycloalkyl groups having 3 to 6 carbon atoms, alkoxy groups having 1 to 6 carbon atoms, aryloxy groups having 5 to 18 carbon atoms, aralkyloxy groups having 7 to 18 carbon atoms, arylamino groups having 5 to 16 carbon atoms, nitro group, cyano group, ester groups having 1 to 6 carbon atoms and halogen atoms. Examples of the above groups and 20 atoms include the atoms and groups described as the examples of the atoms and the groups represented by A<sup>12</sup> to A<sup>14</sup> in general formula (IV) described in the following.

In the present invention, the compound of component (B) is at least one compound selected from [1] anthracene derivatives represented by the 25 following general formula (I), [2] anthracene derivatives represented by the following general formula (II), [3] spirofluorene derivatives

represented by the following general formula (III), [4] compounds having condensed rings represented by the following general formula (IV) and [5] metal complex compounds shown in the following.

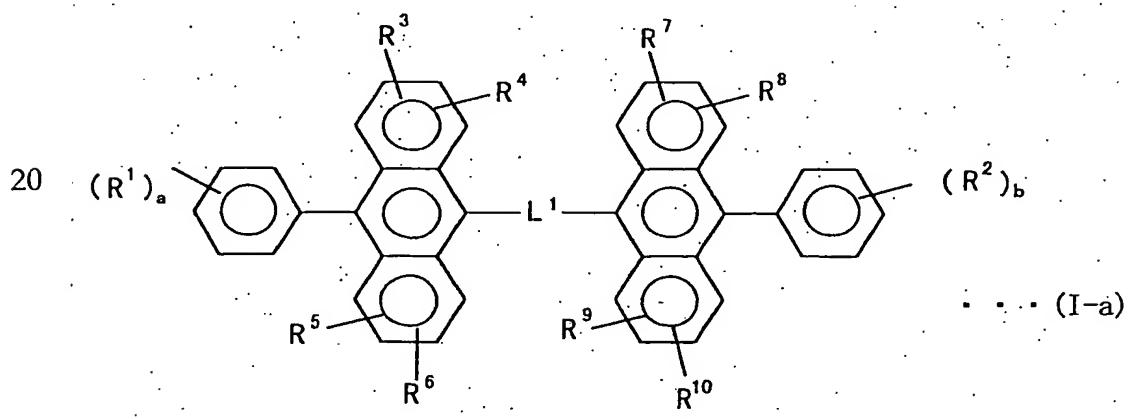
It is preferable that the compound of component (B) is at least one compound selected from the anthracene derivatives represented by general formulae (I) and (II).

[1] Anthracene derivatives represented by general formula (I):



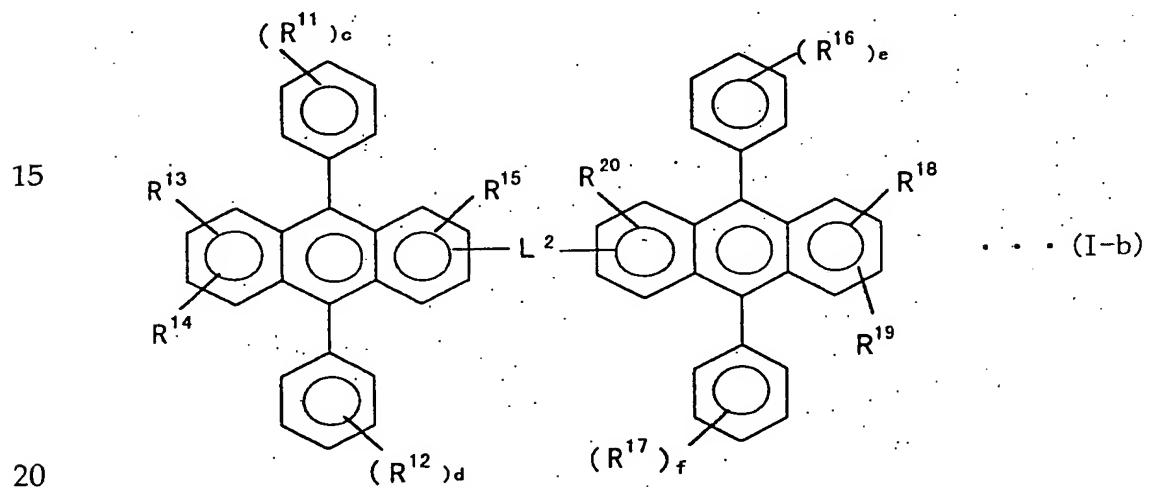
wherein  $A^1$  and  $A^2$  each independently represent a substituted or unsubstituted monophenylanthryl group or a substituted or unsubstituted diphenylanthryl group and may represent the same group or different groups, and  $L$  represents a single bond or a divalent bonding group.

As the anthracene derivative represented by general formula (I), anthracene derivatives represented by the following general formula (I-a) and anthracene derivatives represented by the following general formula (I-b) are preferable.



25 In the above general formula (I-a),  $R^1$  to  $R^{10}$  each independently represent hydrogen atom, an alkyl group, a cycloalkyl group, an aryl group

which may be substituted, an alkoxy group, an aryloxy group, an alkylamino group, an alkenyl group, an arylamino group or a heterocyclic group which may be substituted, a and b each represent an integer of 1 to 5, atoms or groups represented by a plurality of R<sup>1</sup> and R<sup>2</sup> may be the same with or different from each other and may be bonded to each other to form a ring when a and b each represent an integer of 2 or greater, groups represented by combinations of R<sup>3</sup> and R<sup>4</sup>, R<sup>5</sup> and R<sup>6</sup>, R<sup>7</sup> and R<sup>8</sup>, and R<sup>9</sup> and R<sup>10</sup> may be bonded to each other to form a ring, and L<sup>1</sup> represents a single bond, -O-, -S-, -N(R)- (R representing an alkyl group or an aryl group which may be substituted), an alkylene group or an arylene group.



In the above general formula (I-b), R<sup>11</sup> to R<sup>20</sup> each independently represent hydrogen atom, an alkyl group, a cycloalkyl group, an aryl group which may be substituted, an alkoxy group, an aryloxy group, an alkylamino group, an arylamino group or a heterocyclic group which may

be substituted, c, d, e and f each represent an integer of 1 to 5, atoms or groups represented by a plurality of R<sup>11</sup>, R<sup>12</sup>, R<sup>16</sup> and R<sup>17</sup> may be the same with or different from each other and may be bonded to each other to form a ring when c, d, e and f each represent an integer of 2 or greater, groups represented by combinations of R<sup>13</sup> and R<sup>14</sup>, and R<sup>18</sup> and R<sup>19</sup> may be bonded to each other to form a ring, and L<sup>2</sup> represents a single bond, -O-, -S-, -N(R)- (R representing an alkyl group or an aryl group which may be substituted), an alkylene group or an arylene group.

That a group may be substituted means the group is substituted or unsubstituted.

In the above general formulae (I-a) and (I-b), among the groups represented by R<sup>1</sup> to R<sup>20</sup>, alkyl groups having 1 to 6 carbon atoms, cycloalkyl groups having 3 to 6 carbon atoms, aryl groups having 5 to 18 carbon atoms, alkoxy groups having 1 to 6 carbon atoms, aryloxy groups having 5 to 18 carbon atoms and alkenyl groups having 1 to 6 carbon atoms are preferable; amino groups substituted with an aryl group having 5 to 16 carbon atoms are preferable as the arylamino group; and triazole group, oxadiazole group, quinoxaline group, furanyl group and thienyl group are preferable as the heterocyclic groups.

As the alkyl group represented by R in -N(R)- which is represented by L<sup>1</sup> and L<sup>2</sup>, alkyl groups having 1 to 6 carbon atoms, alkylene groups having 1 to 20 carbon atoms and aryl groups having 5 to 18 carbon atoms are preferable.

[2] Anthracene derivatives represented by general formula (II):

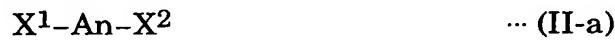
A<sup>3</sup>-An-A<sup>4</sup> ... (II)

wherein An represents a substituted or unsubstituted divalent anthracene

residue group,  $A^3$  and  $A^4$  each independently represent a substituted or unsubstituted aryl group having 6 to 40 carbon atoms, at least one of  $A^3$  and  $A^4$  represents a substituted or unsubstituted monovalent condensed aromatic ring or a substituted or unsubstituted aryl group having 10 or 5 more carbon atoms, and  $A^3$  and  $A^4$  may represent the same group or different groups.

Examples of the aryl group represented by  $A^3$  and  $A^4$  include phenyl group, 2-methylphenyl group, 3-methylphenyl group, 4-methylphenyl group, 4-ethylphenyl group, biphenyl group, 4-methylbiphenyl group, 4-10 ethylbiphenyl group, 4-cyclohexylbiphenyl group, terphenyl group, 3,5-dichlorophenyl group and condensed aromatic ring groups which are residue groups derived from naphthalene, phenanthrene, fluoranthene, anthracene, pyrene, perylene, coronene, chrysene, picene, fluorene, terphenyl, diphenylanthracene, biphenyl, carbazole, triphenylene, 15 rubicene, benzoanthracene, phenylanthracene, bisanthracene, dianthracylbenzene and dibenzoanthracene, which may be substituted or unsubstituted.

As the anthracene derivative represented by the above general formula (II), anthracene derivatives represented by the following general 20 formula (II-a):

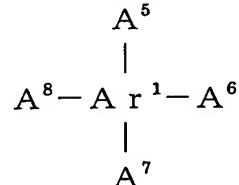


wherein  $An$  represents a substituted or unsubstituted divalent anthracene residue group and  $X^1$  and  $X^2$  each independently represent a monovalent residue group derived from naphthalene, phenanthrene, fluoranthene, 25 anthracene, pyrene, perylene, coronene, chrysene, picene, diphenylanthracene, carbazole, triphenylene, rubicene, benzoanthracene,

phenylanthracene, bisanthracene, dianthracenylbenzene or dibenzoanthracene, which may be substituted or unsubstituted, are preferable.

[3] Spirofluorene derivatives represented by general formula (III):

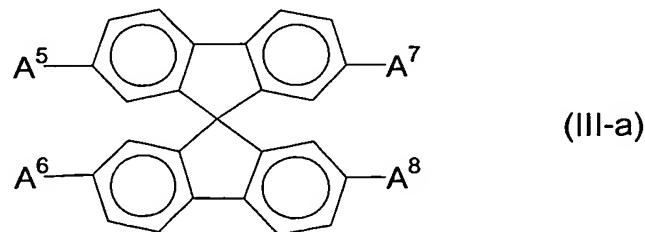
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10 wherein  $A r^1$  represents a substituted or unsubstituted spirofluorene residue group,  $A^5$  to  $A^8$  each independently represent a substituted or unsubstituted aryl group having 6 to 40 carbon atoms.

Examples of the substituted or unsubstituted aryl group represented by  $A^5$  to  $A^8$  in the above general formula (III) include phenyl group, 2-methylphenyl group, 3-methylphenyl group, 4-methylphenyl group, 4-ethylphenyl group, biphenyl group, 4-methylbiphenyl group, 4-ethylbiphenyl group, 4-cyclohexylbiphenyl group, terphenyl group, 3,5-dichlorophenyl group, naphthyl group, 5-methylnaphthyl group, anthryl group and pyrenyl group.

20 As the spirofluorene derivative represented by the above general formula (III), spirofluorene derivatives represented by the following general formula (III-a):

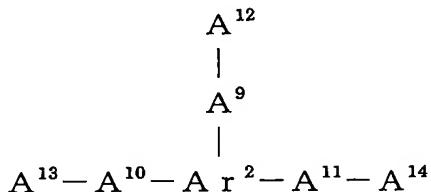


wherein  $A^5$  to  $A^8$  each independently represent a substituted or

unsubstituted biphenyl group or a substituted or unsubstituted naphthyl group, are preferable.

[4] Compounds having condensed rings represented by general formula (IV):

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10 wherein  $Ar^2$  represents a substituted or unsubstituted aromatic ring group having 6 to 40 carbon atoms,  $A^9$  to  $A^{11}$  each independently represent a substituted or unsubstituted arylene group having 6 to 40 carbon atoms,  $A^{12}$  to  $A^{14}$  each independently represent hydrogen atom, an alkyl group having 1 to 6 carbon atoms, a cycloalkyl group having 3 to 6 carbon atoms, 15 an alkoxy group having 1 to 6 carbon atoms, an aryloxy group having 5 to 18 carbon atoms, an aralkyloxy group having 7 to 18 carbon atoms, an arylamino group having 5 to 16 carbon atoms, nitro group, cyano group, an ester group having 1 to 6 carbon atoms or a halogen atom, and at least one of  $A^9$  to  $A^{14}$  represents a group having condensed aromatic rings.

20 Examples of the aromatic ring group represented by  $Ar^2$  include residue groups derived from benzene, biphenyl, terphenyl, phenanthrene, fluoranthene, anthracene, pyrene, perylene, coronene, chrysene, picene, fluorene, carbazole, rubicene, benzoanthracene and dibenzoanthracene.

25 Examples of the arylene group represented by  $A^9$  to  $A^{11}$  include divalent residue groups derived from the aromatic compounds described above as the examples of the aromatic ring group represented by  $Ar^2$ .

Examples of the alkyl group having 1 to 6 carbon atoms represented by  $A^{12}$  to  $A^{14}$  include methyl group, ethyl group, propyl group, isopropyl

group, butyl group, isobutyl group, sec-butyl group, tert-butyl group, various types of pentyl group and various types of hexyl group.

Examples of the cycloalkyl group having 3 to 6 carbon atoms represented by A<sup>12</sup> to A<sup>14</sup> include cyclopropyl group, cyclobutyl group, 5 cyclopentyl group and cyclohexyl group.

Examples of the alkoxy group having 1 to 6 carbon atoms represented by A<sup>12</sup> to A<sup>14</sup> include methoxyl group, ethoxyl group, propoxyl group, isopropoxyl group, butoxyl group, isobutoxyl group, sec-butoxyl group, tert-butoxyl group, various types of pentyloxy groups and various 10 types of hexyloxy groups.

Examples of the aryloxy group having 5 to 18 carbon atoms represented by A<sup>12</sup> to A<sup>14</sup> include phenoxy group, tolyloxy group and naphthoxy group.

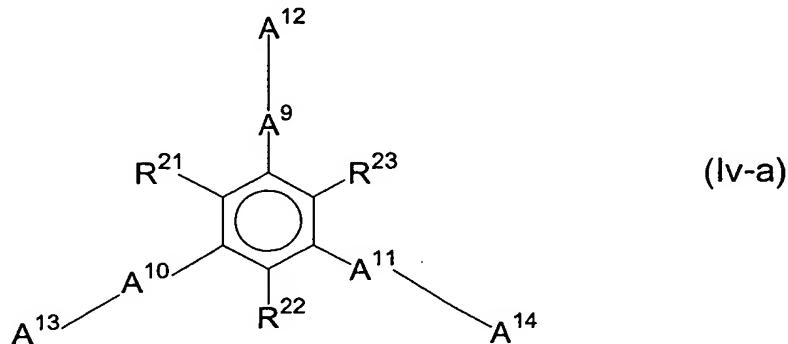
Examples of the aralkyloxy group having 7 to 18 carbon atoms 15 represented by A<sup>12</sup> to A<sup>14</sup> include benzyloxy group, phenyloxy group and naphthylmethoxyl group.

Examples of the arylamino group having 5 to 16 carbon atoms represented by A<sup>12</sup> to A<sup>14</sup> include diphenylamino group, ditolylamino group, dinaphthylamino group and naphthylphenylamino group.

20 Examples of the ester group having 1 to 6 carbon atoms represented by A<sup>12</sup> to A<sup>14</sup> include methoxycarbonyl group, ethoxycarbonyl group, propoxycarbonyl group and isopropoxycarbonyl group.

Examples of the halogen atoms represented by A<sup>12</sup> to A<sup>14</sup> include 25 fluorine atom, chlorine atom and bromine atom. The aryl group in the present invention includes styrylphenyl group, styrylbiphenyl group and styrylnaphthyl group.

As the compound having condensed rings represented by general formula (IV), compounds having condensed rings represented by the following general formula (IV-a):



wherein A<sup>9</sup> to A<sup>14</sup> are as defined above, R<sup>21</sup> to R<sup>23</sup> each independently  
5 represent hydrogen atom, an alkyl group having 1 to 6 carbon atoms, a  
cycloalkyl group having 3 to 6 carbon atoms, an alkoxy group having 1 to 6  
carbon atoms, an aryloxy group having 5 to 18 carbon atoms, an aralkyloxy group having  
10 7 to 18 carbon atoms, an arylamino group having 5 to 16 carbon atoms, nitro group, cyano group, an ester group having 1 to  
15 6 carbon atoms or a halogen atom, and at least one of A<sup>9</sup> to A<sup>14</sup> represents  
a group having condensed aromatic rings having at least 3 rings, are  
preferable.

Examples of the groups represented by R<sup>21</sup> to R<sup>23</sup> include the  
groups described as the examples of the groups represented by A<sup>12</sup> to A<sup>14</sup>  
15 in general formula (IV).

Examples of the substituent in the compounds represented by the  
above general formulae (I) to (IV), (I-a), (I-b), (II-a), (III-a) and (IV-a)  
include alkyl groups having 1 to 6 carbon atoms, cycloalkyl groups having  
3 to 6 carbon atoms, alkoxy groups having 1 to 6 carbon atoms, aryloxy  
20 groups having 5 to 18 carbon atoms, aralkyloxy groups having 7 to 18

carbon atoms, arylamino groups having 5 to 16 carbon atoms, nitro group, cyano group, ester groups having 1 to 6 carbon atoms and halogen atoms. Specific examples of the above substituent include the substituents described as the examples of the substituents for the groups represented by A<sup>12</sup> to A<sup>14</sup> in the above general formula (IV).

### [5] Metal complex compounds

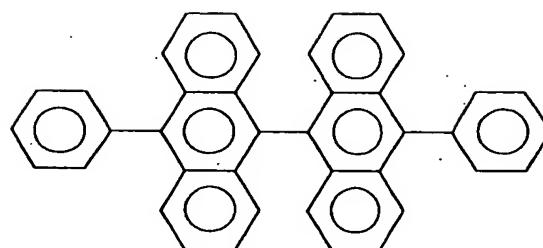
Examples of the metal complex compound described above include 8-hydroxyquinolinatolithium, bis(8-hydroxyquinolinato)zinc, bis(8-hydroxyquinolinato)copper, bis(8-hydroxyquinolinato)manganese, tris(8-hydroxyquinolinato)aluminum, tris(2-methyl-8-hydroxyquinolinato)-aluminum, tris(8-hydroxyquinolinato)gallium, bis(10-hydroxybenzo[h]-quinolinato)beryllium, bis(10-hydroxybenzo[h]quinolinato)zinc, bis(2-methyl-8-hydroxyquinolinato)chlorogallium, bis(2-methyl-8-hydroxy-quinolinato)(o-cresolato)gallium, bis(2-methyl-8-hydroxyquinolinato)-(1-naphtholato)aluminum and bis(2-methyl-8-hydroxyquinolinato)-(2-naphtholate)gallium. Among these compounds, aluminum chelate complex compounds such as tris(8-hydroxyquinolinato)aluminum, tris(2-methyl-8-hydroxyquinolinato)aluminum and bis(2-methyl-8-hydroxyquinolinato)(1-naphtholato)aluminum are preferable.

In the present invention, the anthracene derivative of component (B) may be used singly or in combination of two or more.

Specific examples of the anthracene derivative represented by the above general formula (I-a) are shown in the following.

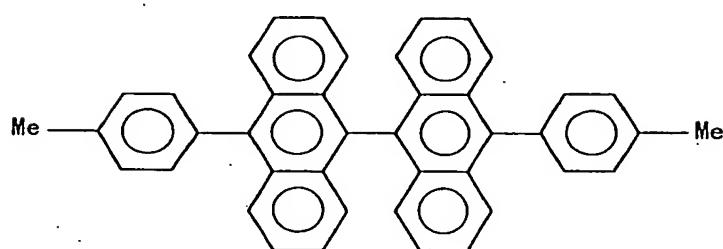
EM1

5



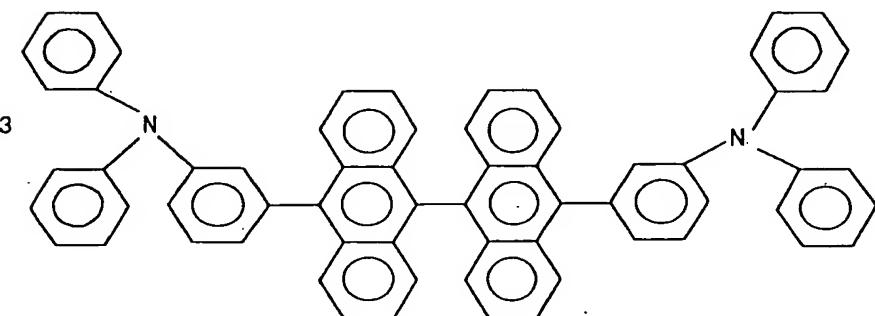
EM2

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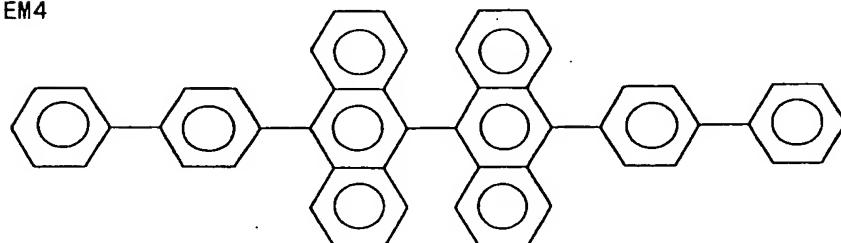


(Me: methyl group; similarly in the following formulae)

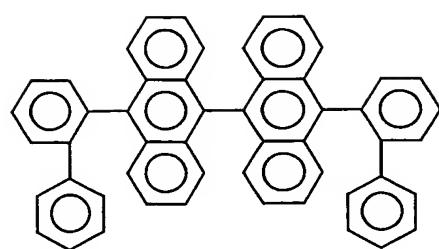
EM3



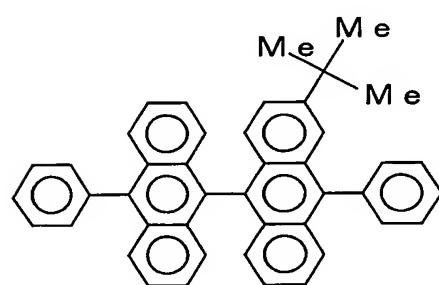
EM4



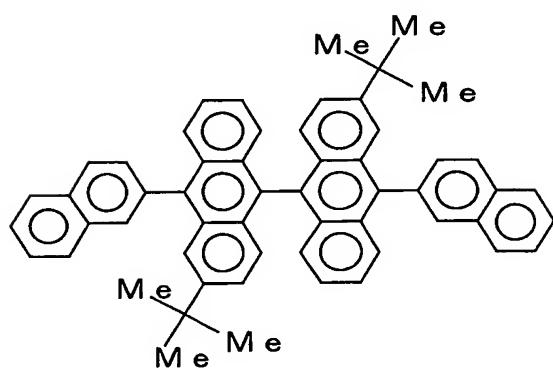
EM 5



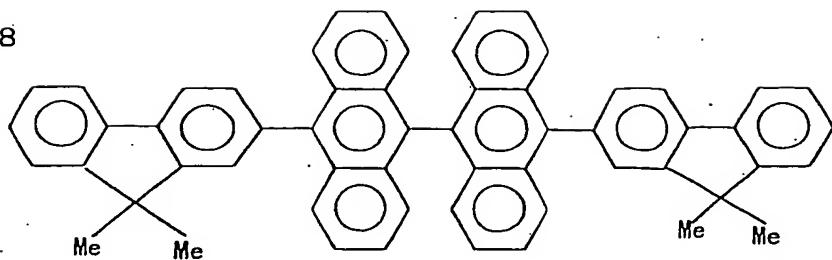
EM 6



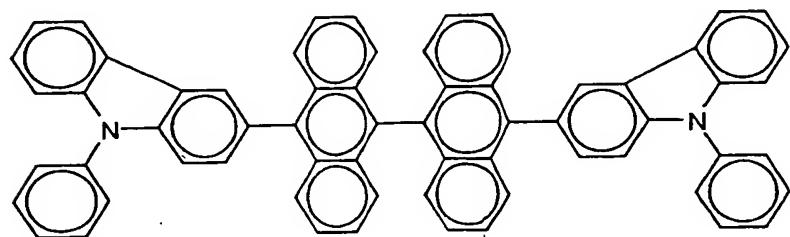
EM 7



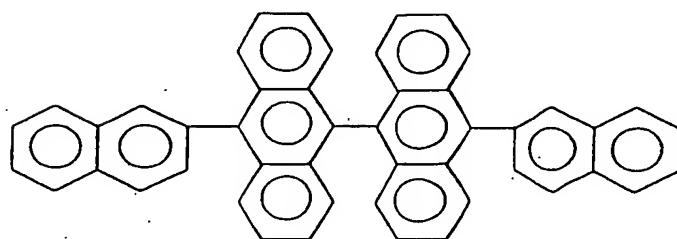
EM8



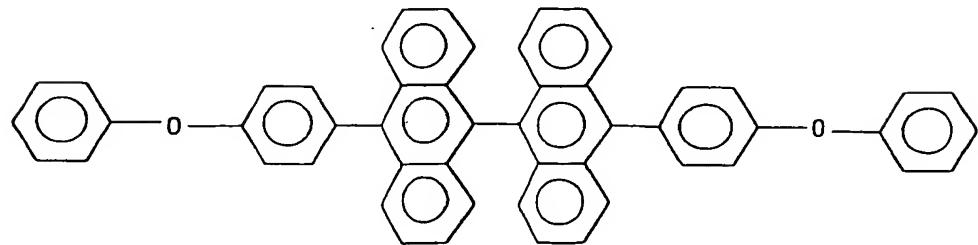
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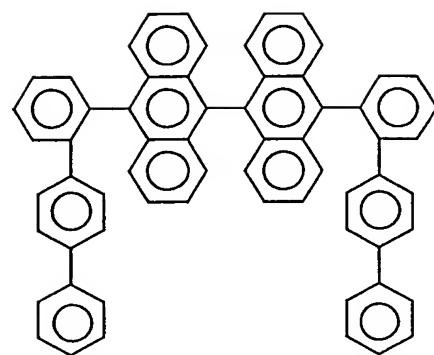
EM10



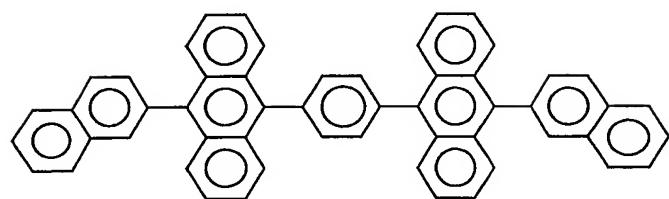
EM11



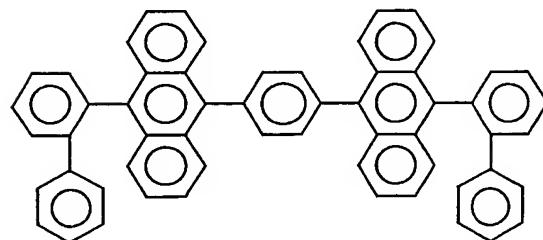
E M12



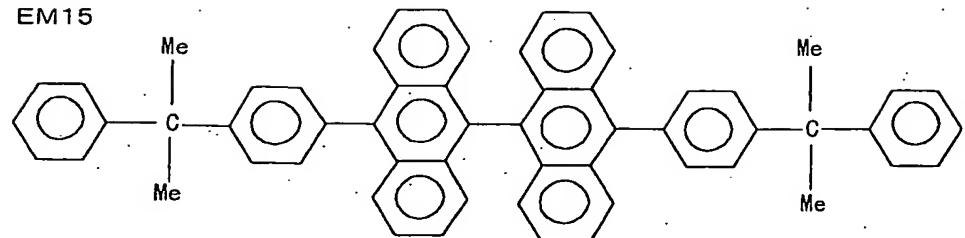
E M13



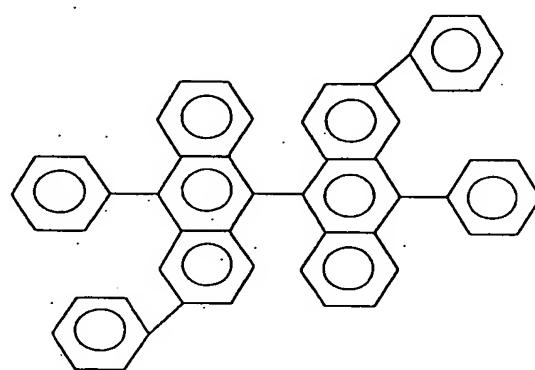
E M14



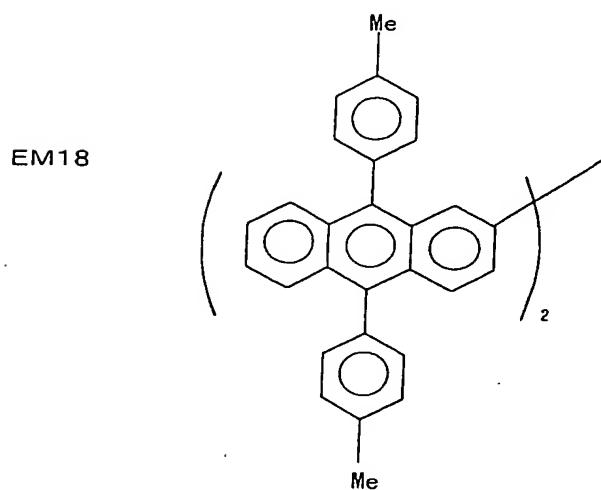
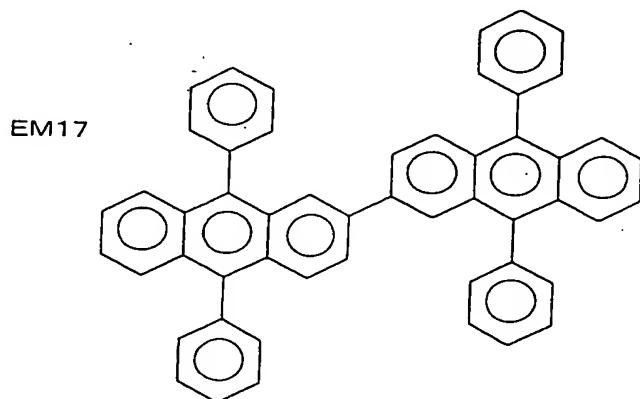
EM15

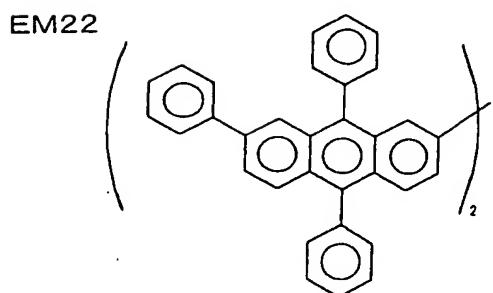
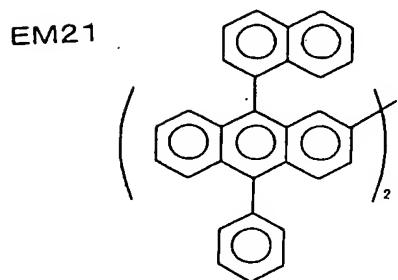
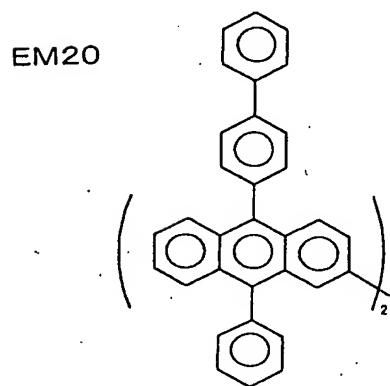
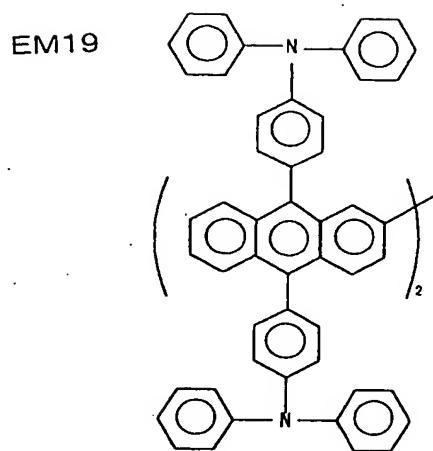


EM16

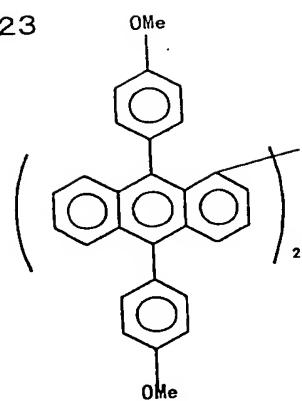


Specific examples of the anthracene derivative represented by the above general formula (I-b) are shown in the following.

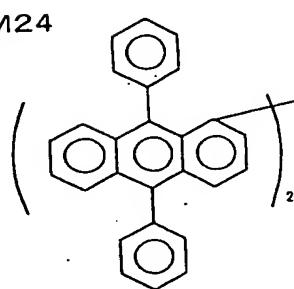




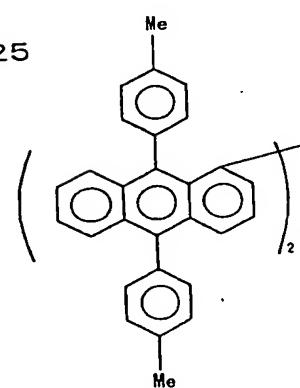
EM23



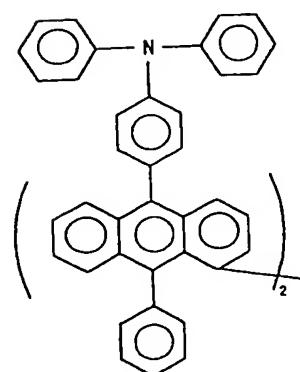
EM24



EM25

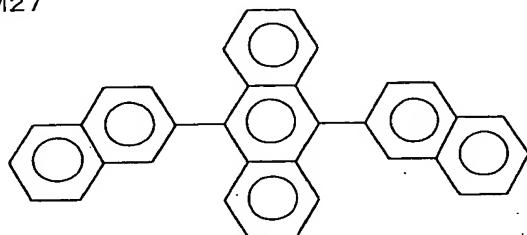


EM26

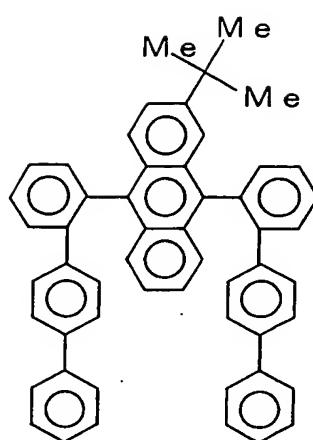


Specific examples of the anthracene derivatives represented by the above general formula (II-a) are shown in the following.

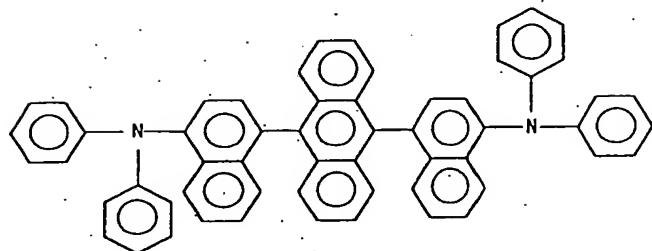
EM27



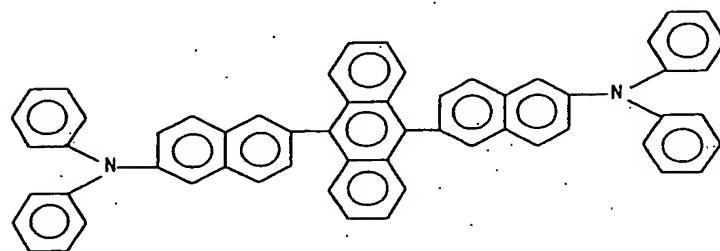
EM32



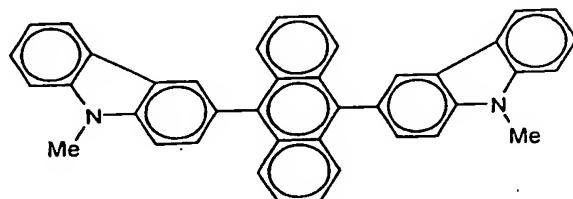
EM33



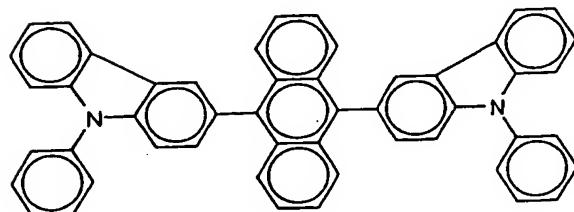
EM34



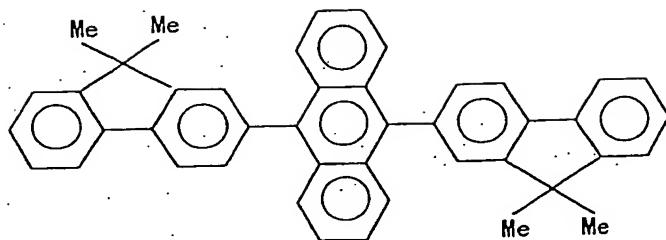
EM35



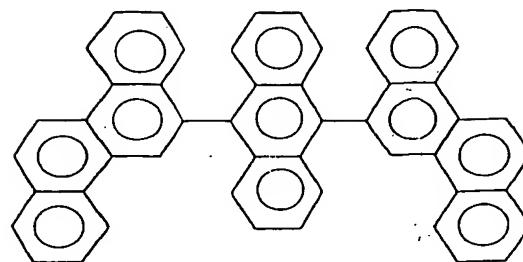
EM36



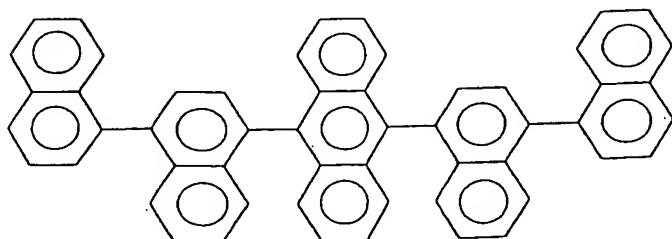
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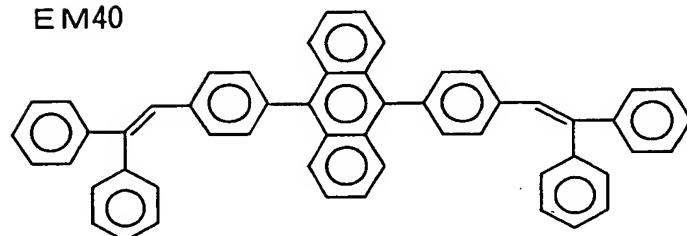
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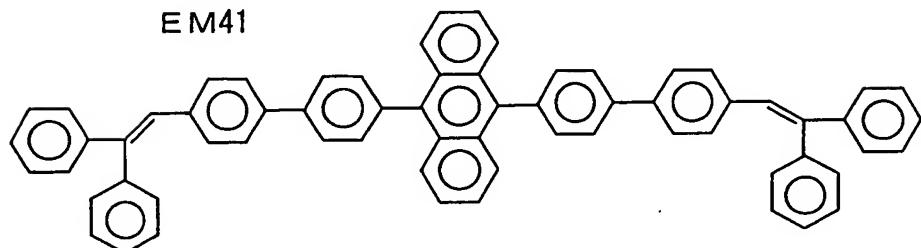
EM39



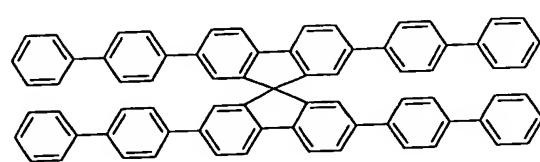
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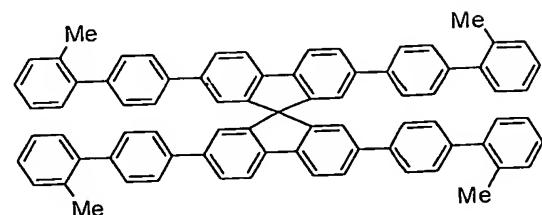
EM41



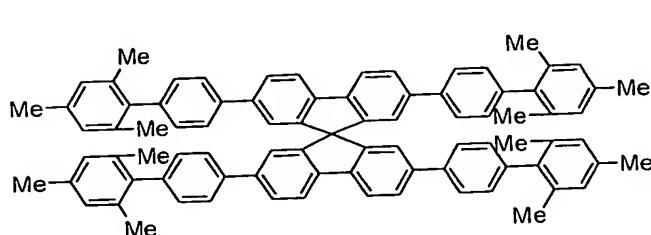
Specific examples of the spirofluorene derivative represented by the above general formula (III-a) are shown in the following.



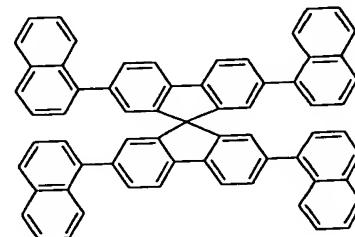
EM42



EM43



EM44

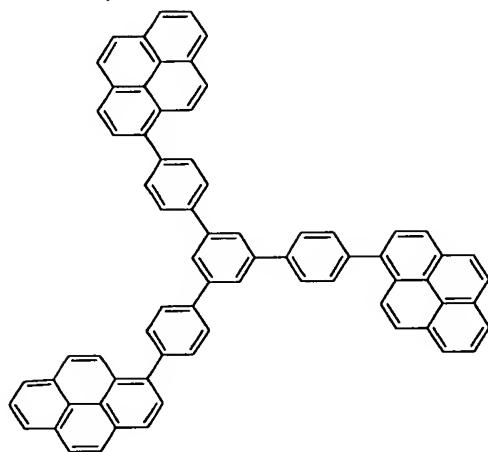


EM45

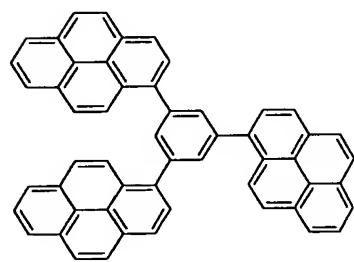
Specific examples of the compound having condensed rings represented by the above general formula (IV-a) are shown in the following.

5

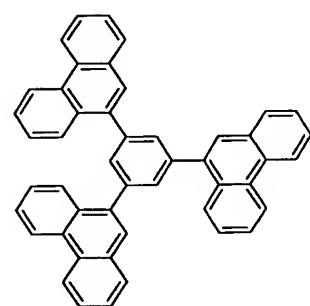
EM49



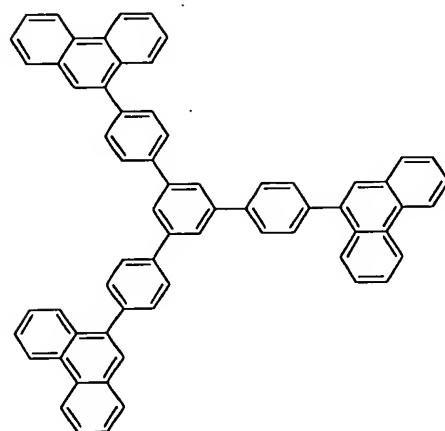
EM46



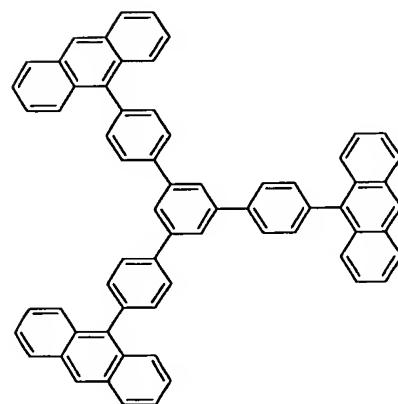
EM47



EM50

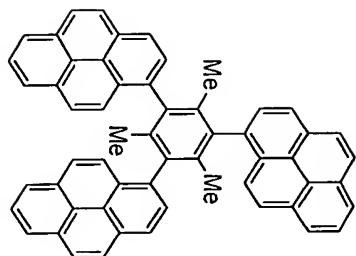


EM48



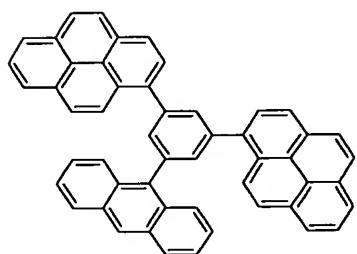
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EM54



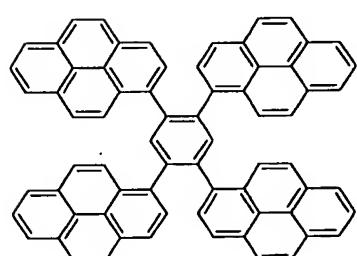
10

EM55



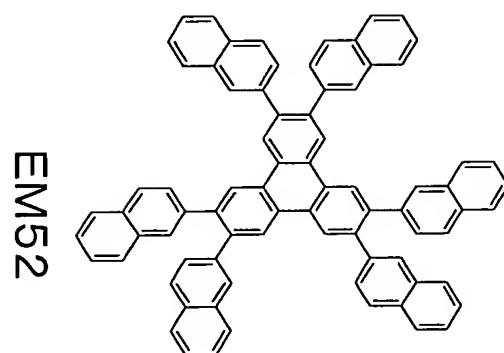
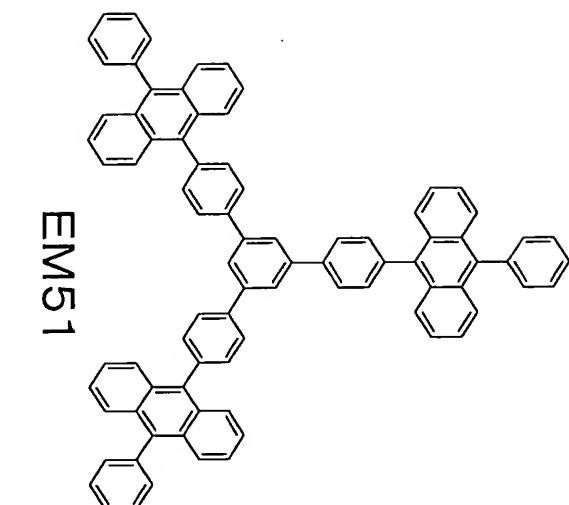
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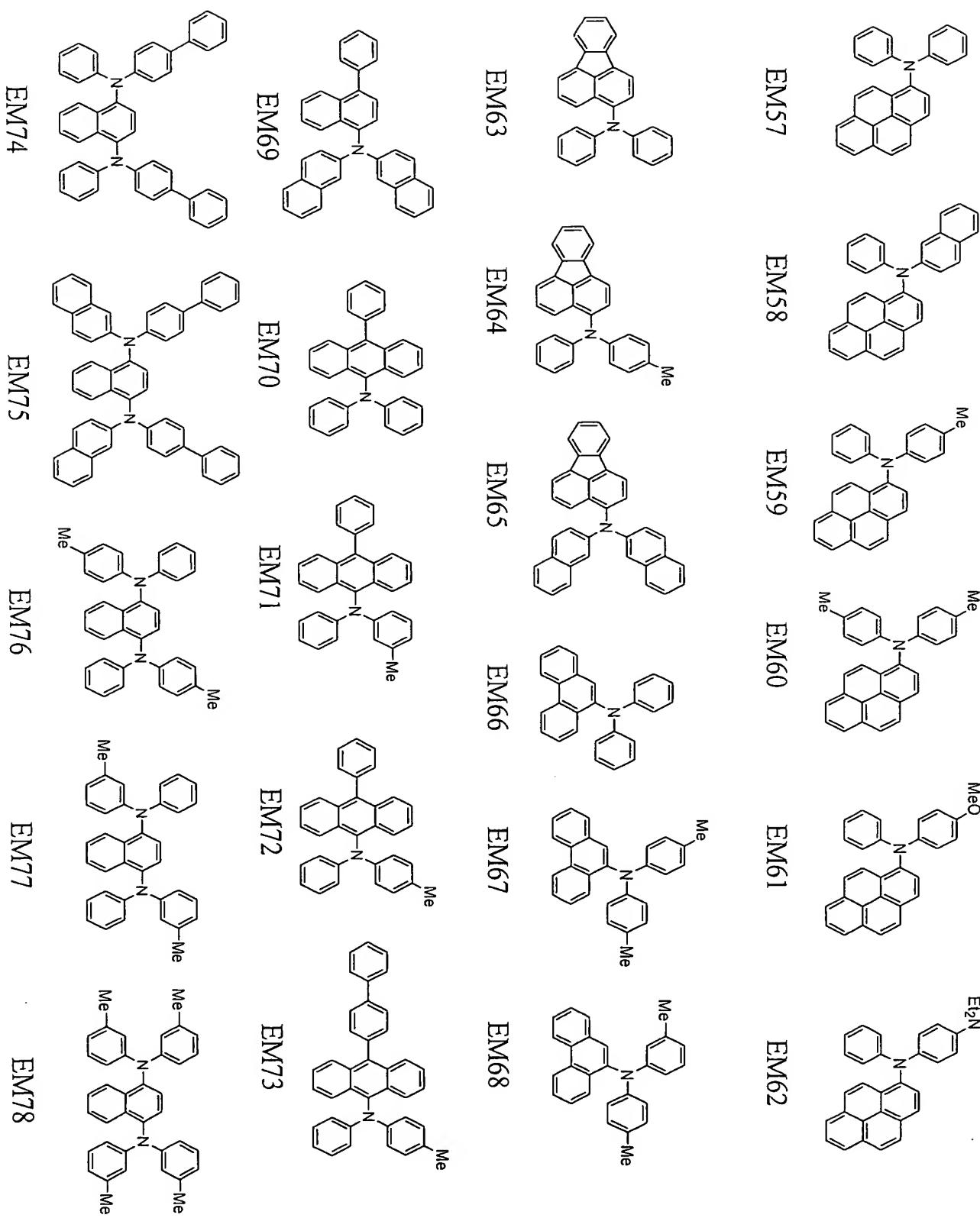
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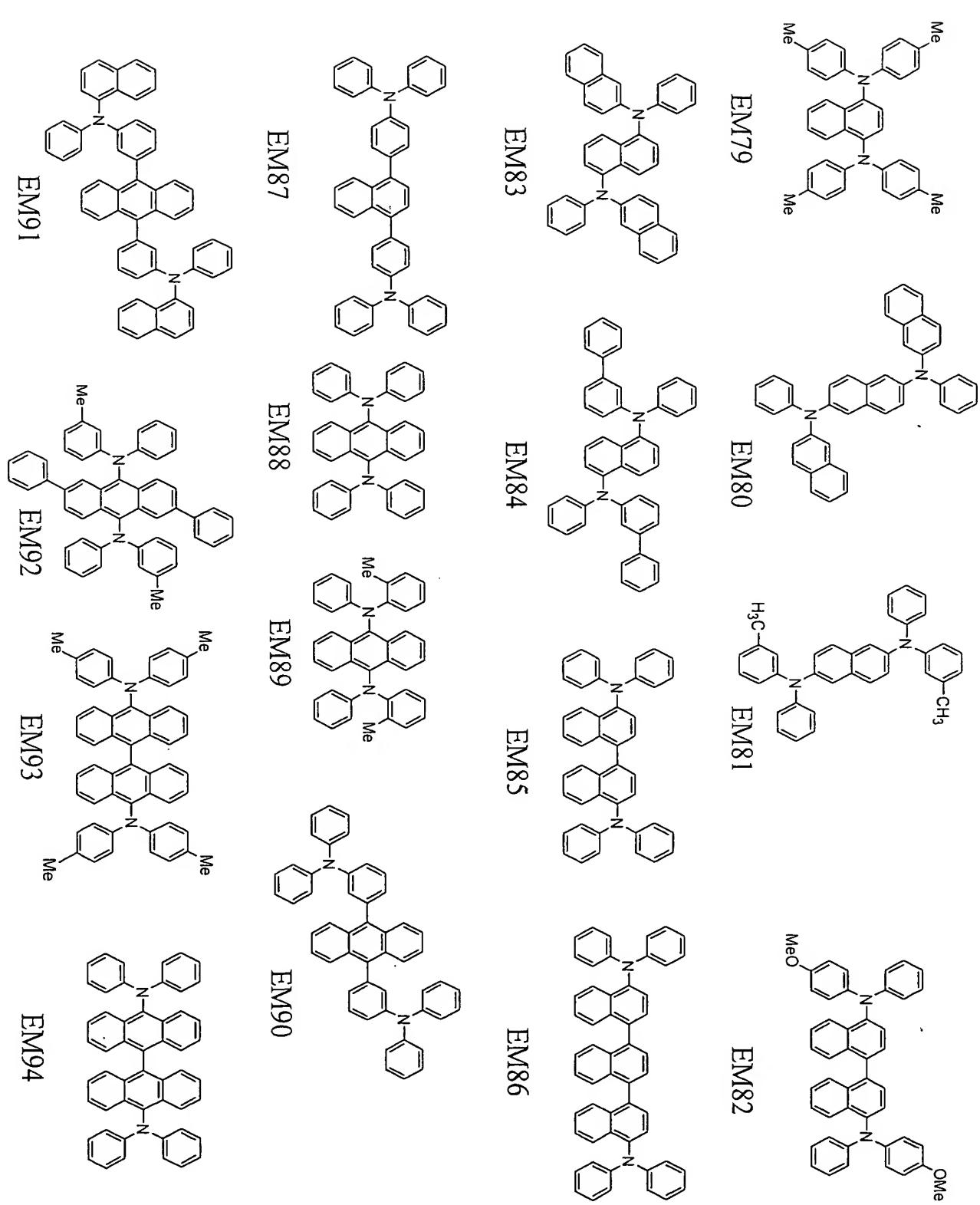


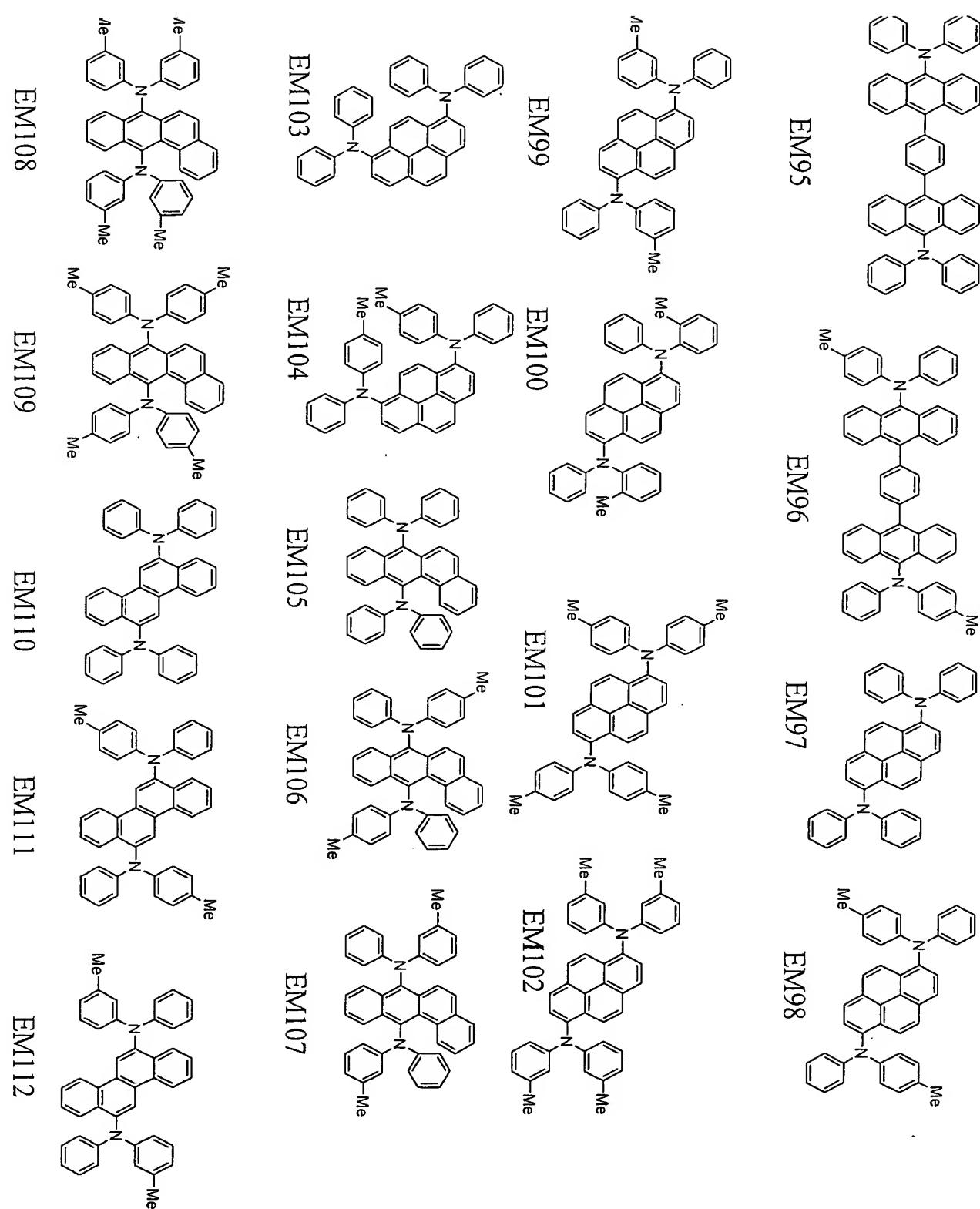
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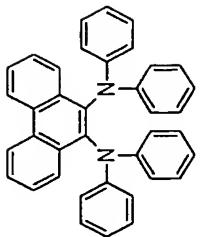
Specific examples of the arylamine compounds represented by the above general formulae (V), (V-a) and (V-b) are shown in the following.



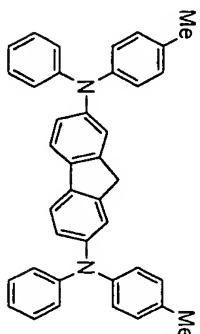




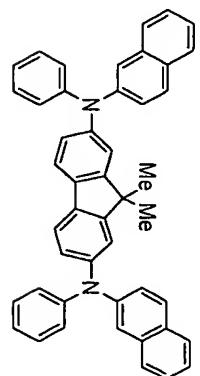




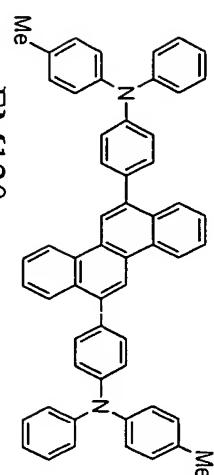
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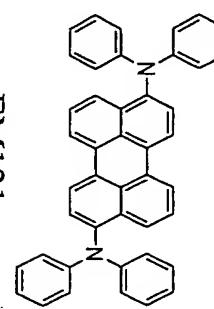
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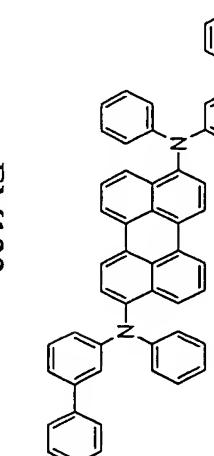
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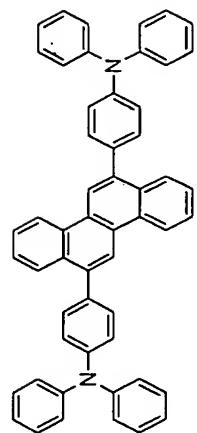
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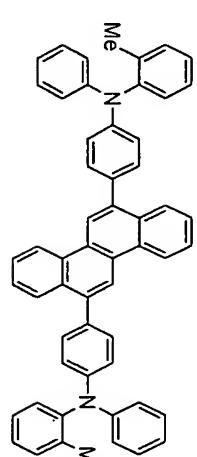
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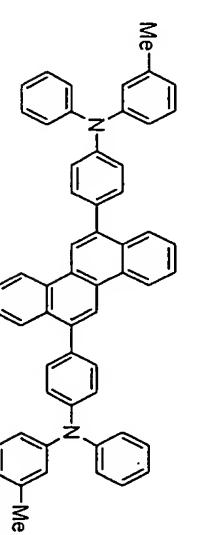
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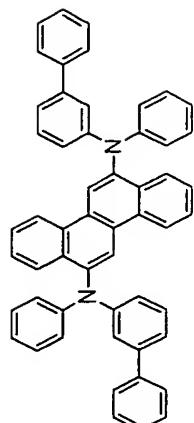
EMI 17



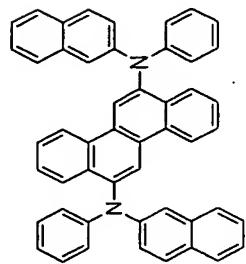
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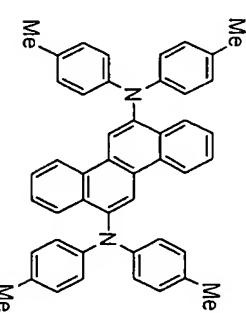
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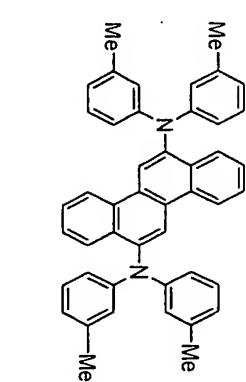
EM113



EM114

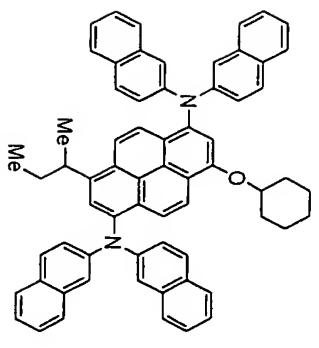


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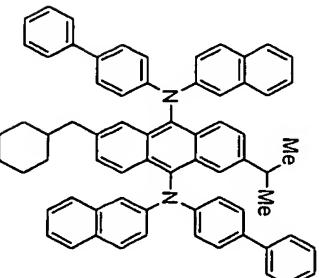


EMI 16

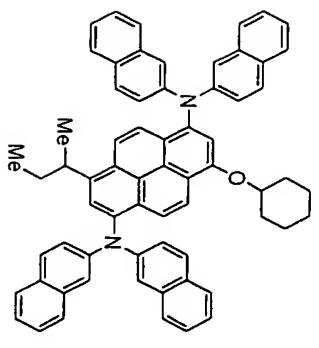
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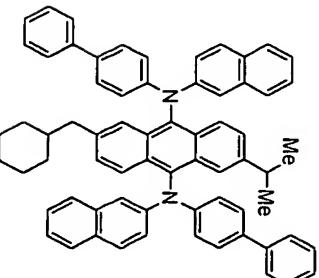
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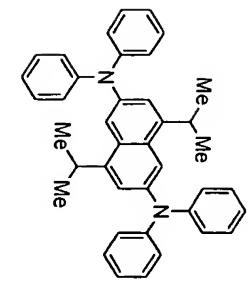
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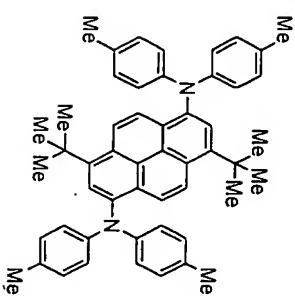
EM137



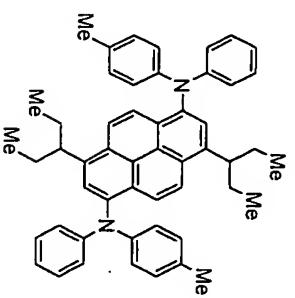
EM130



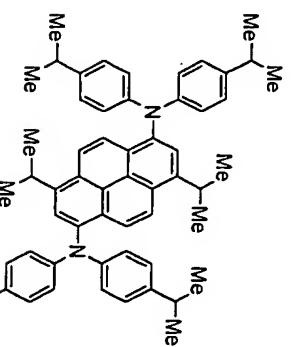
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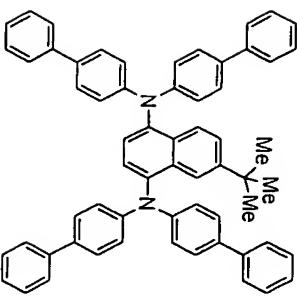
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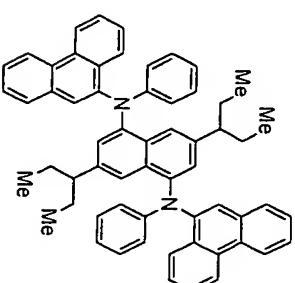
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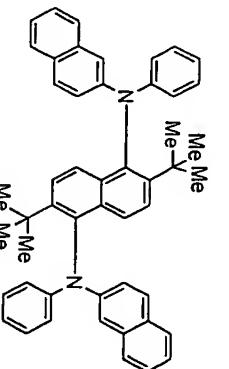
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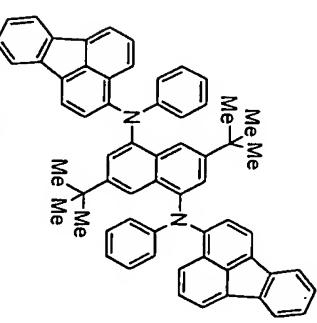
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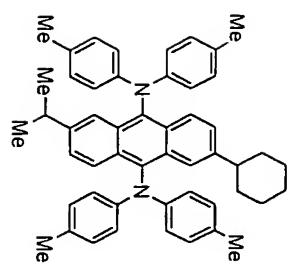


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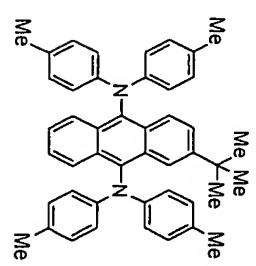


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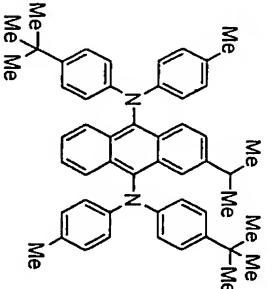




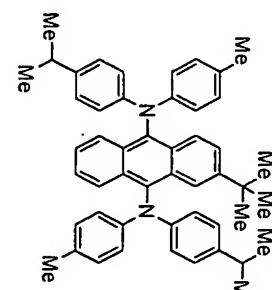
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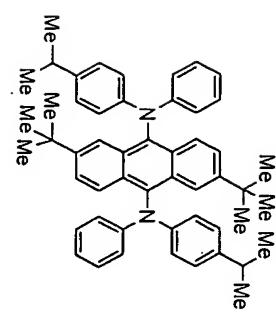
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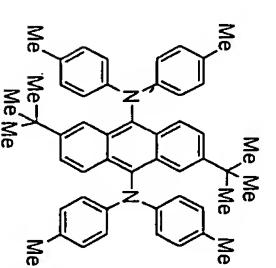
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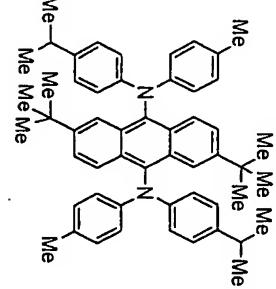
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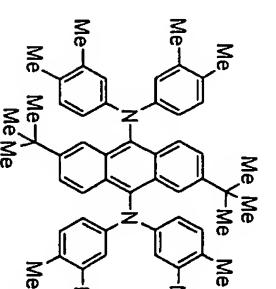
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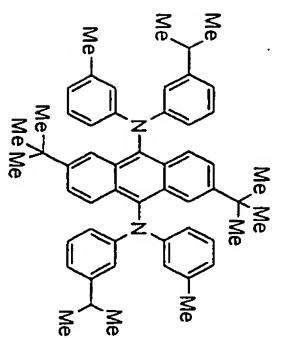
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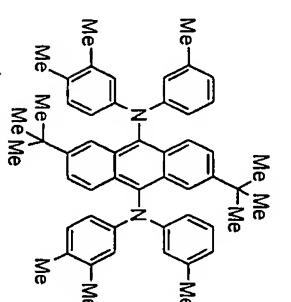
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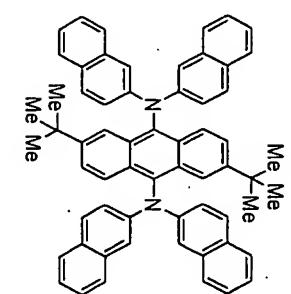
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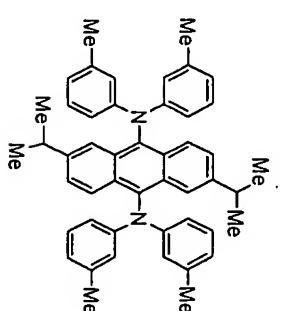
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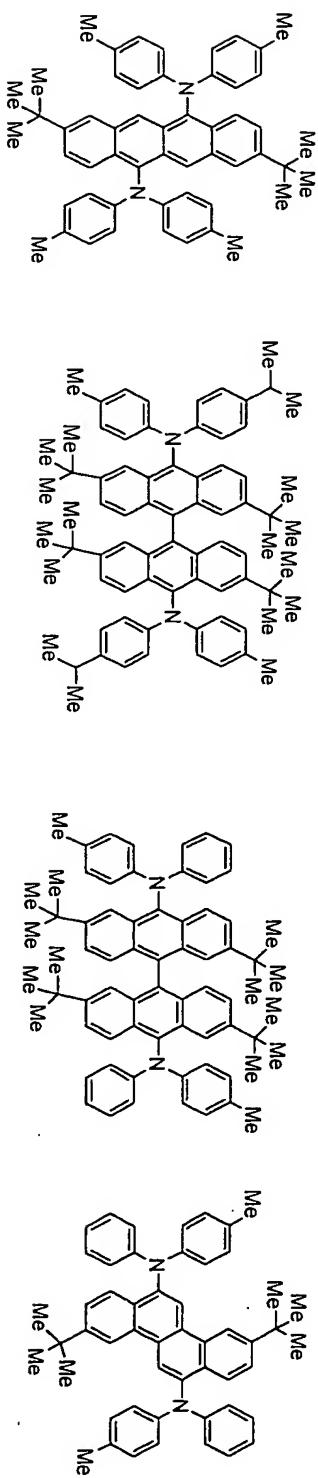
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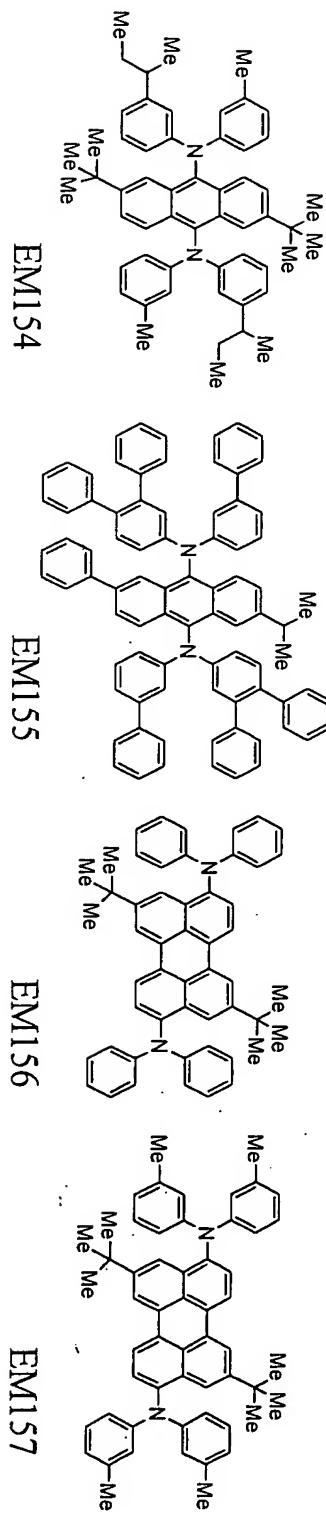


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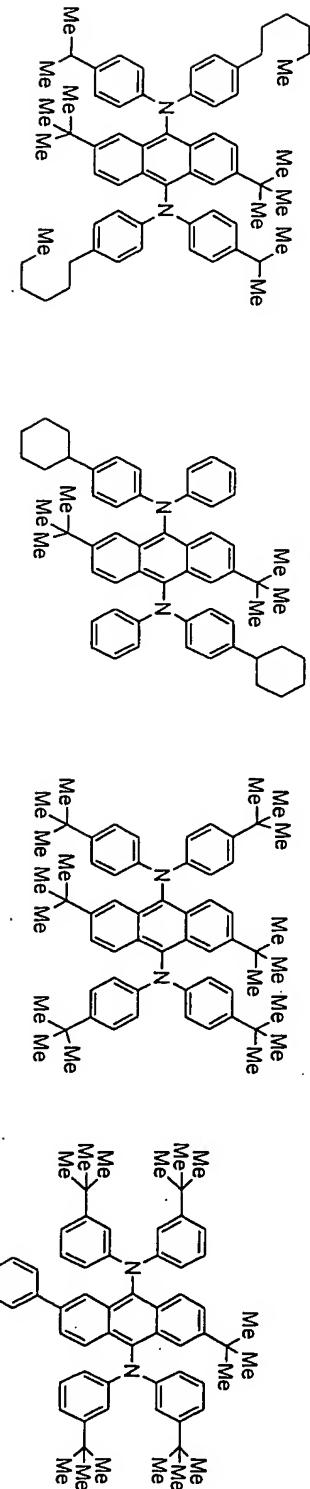
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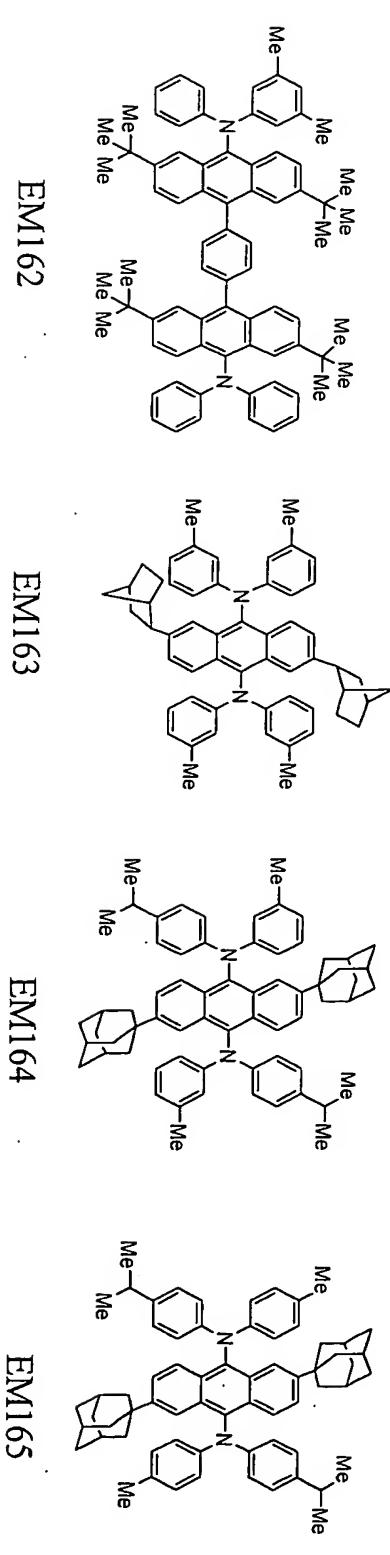
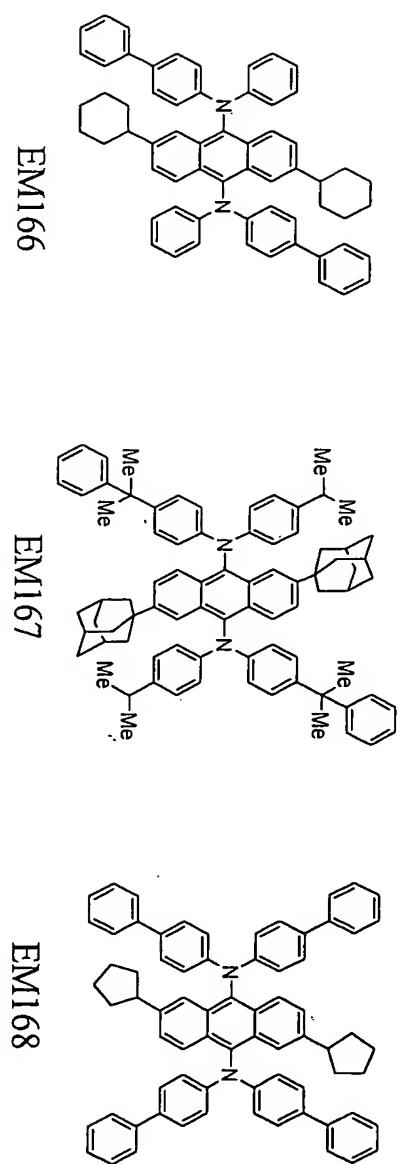
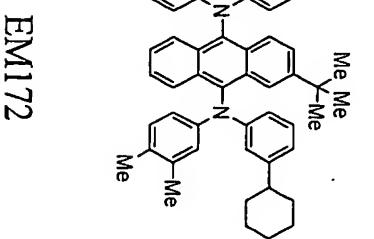
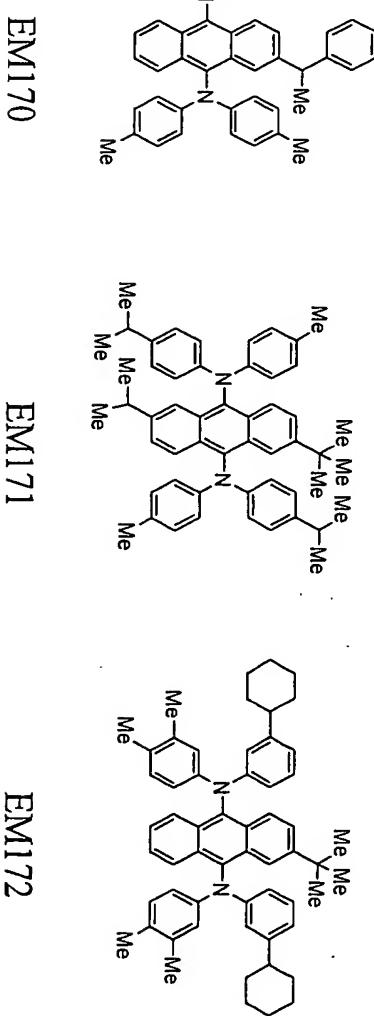
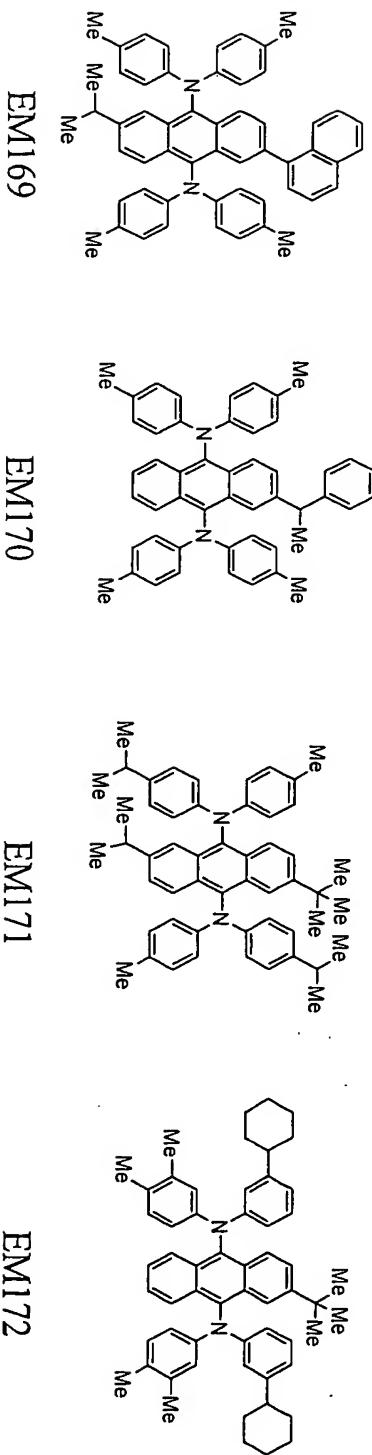
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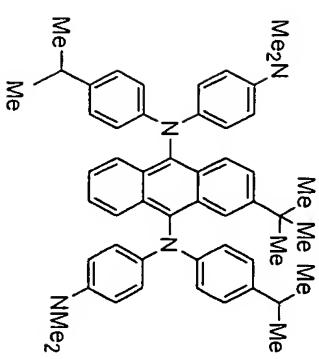
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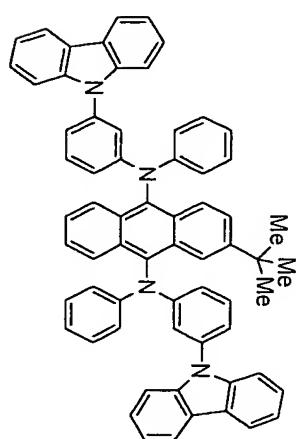
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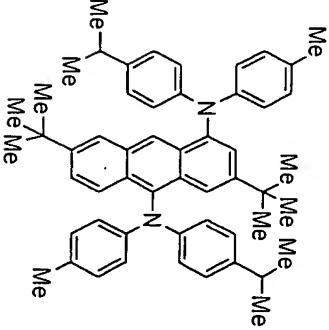




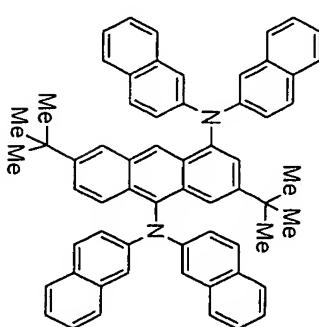
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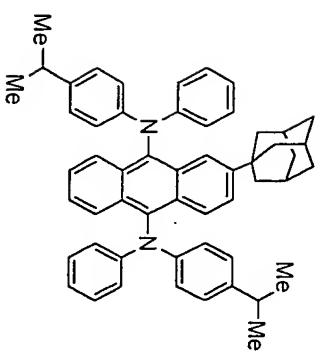
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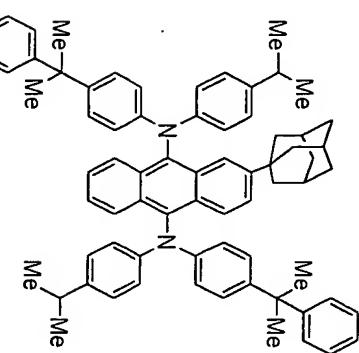
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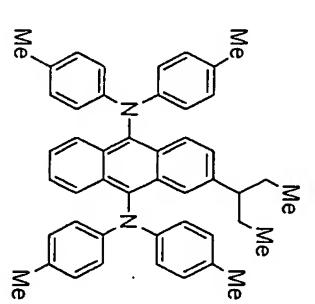
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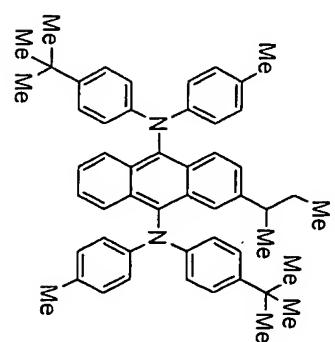
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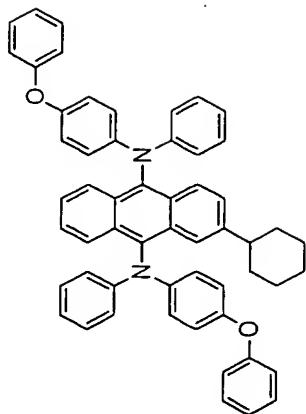
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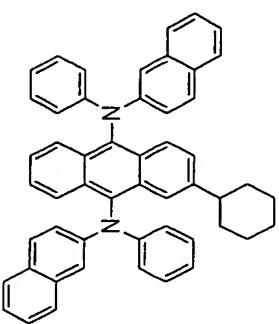
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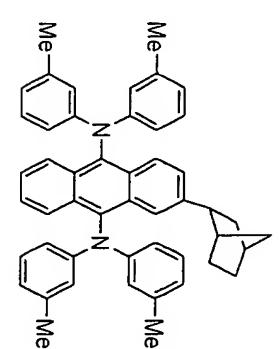
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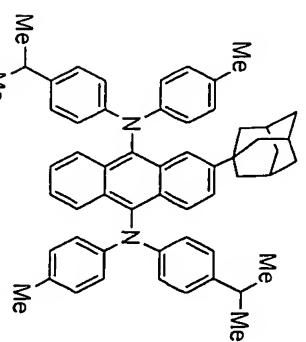
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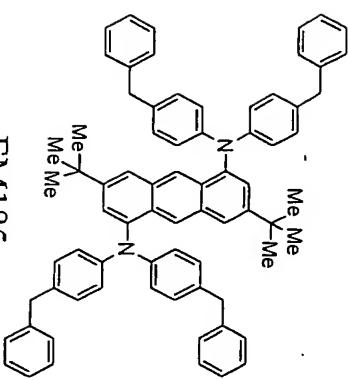
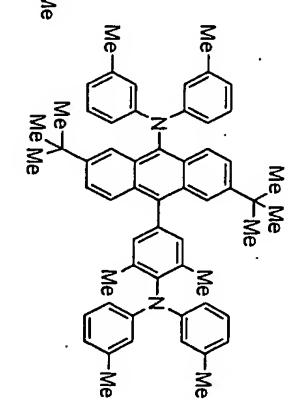
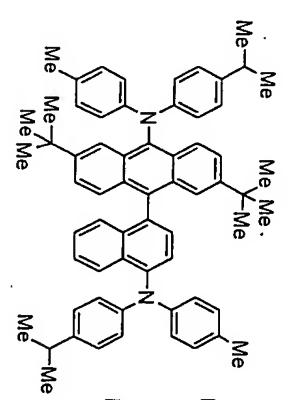
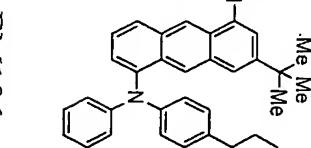
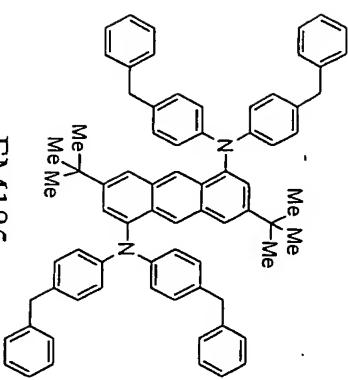
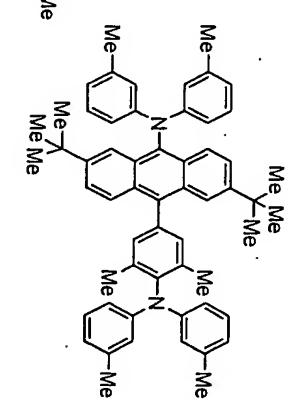
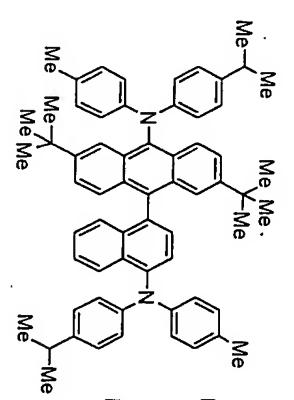
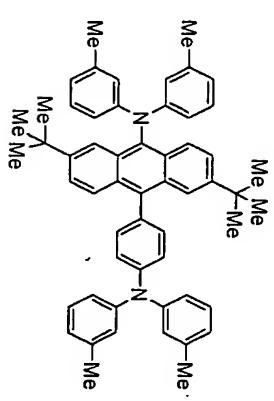
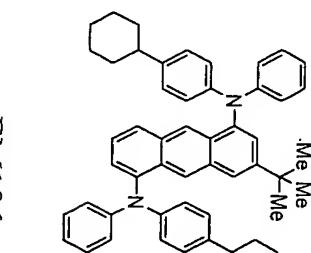
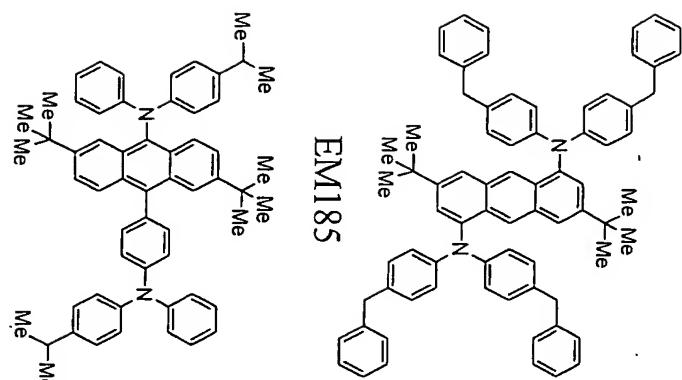
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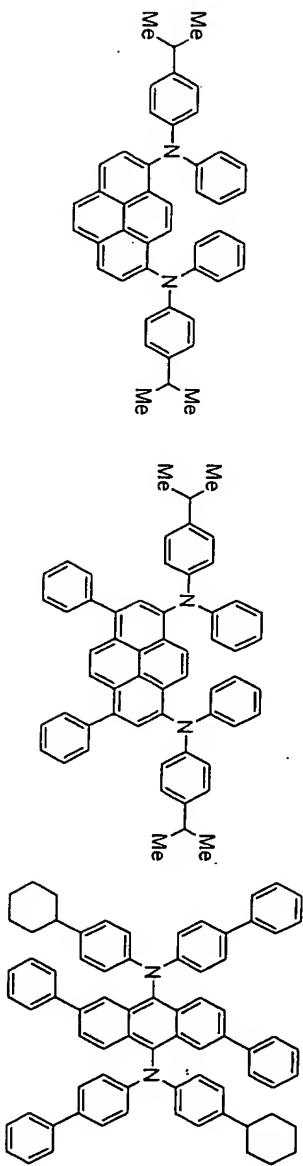


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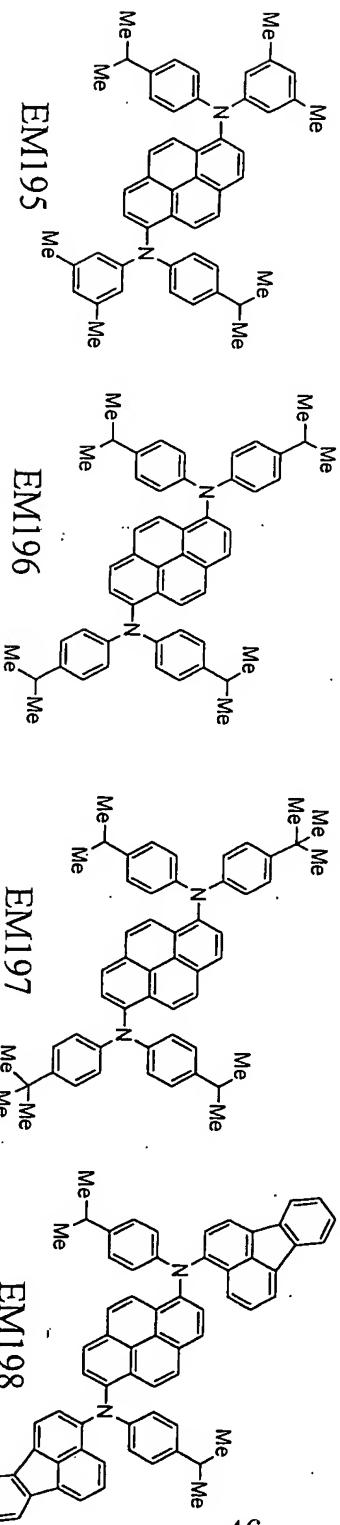




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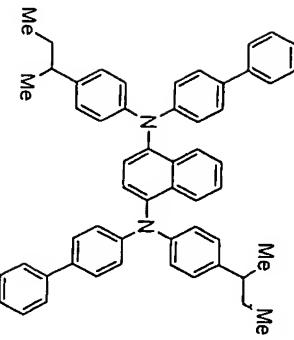


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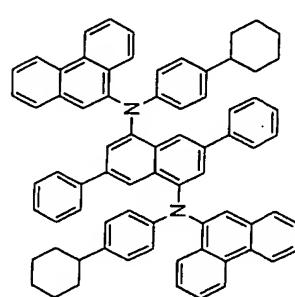
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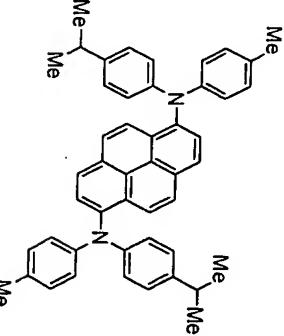
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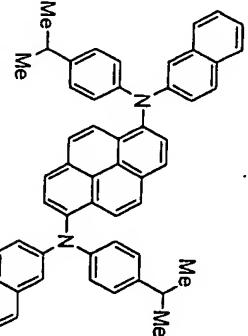
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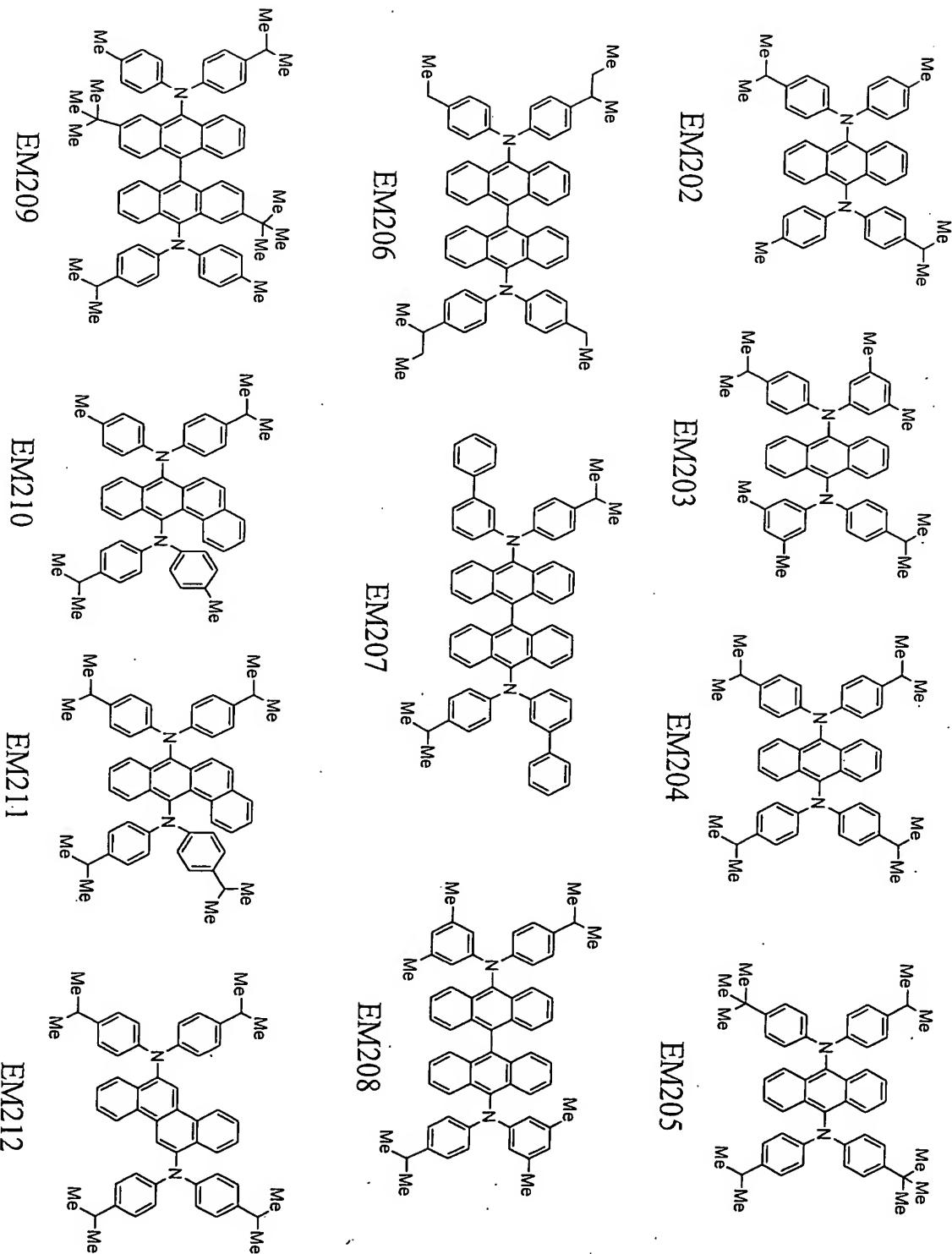
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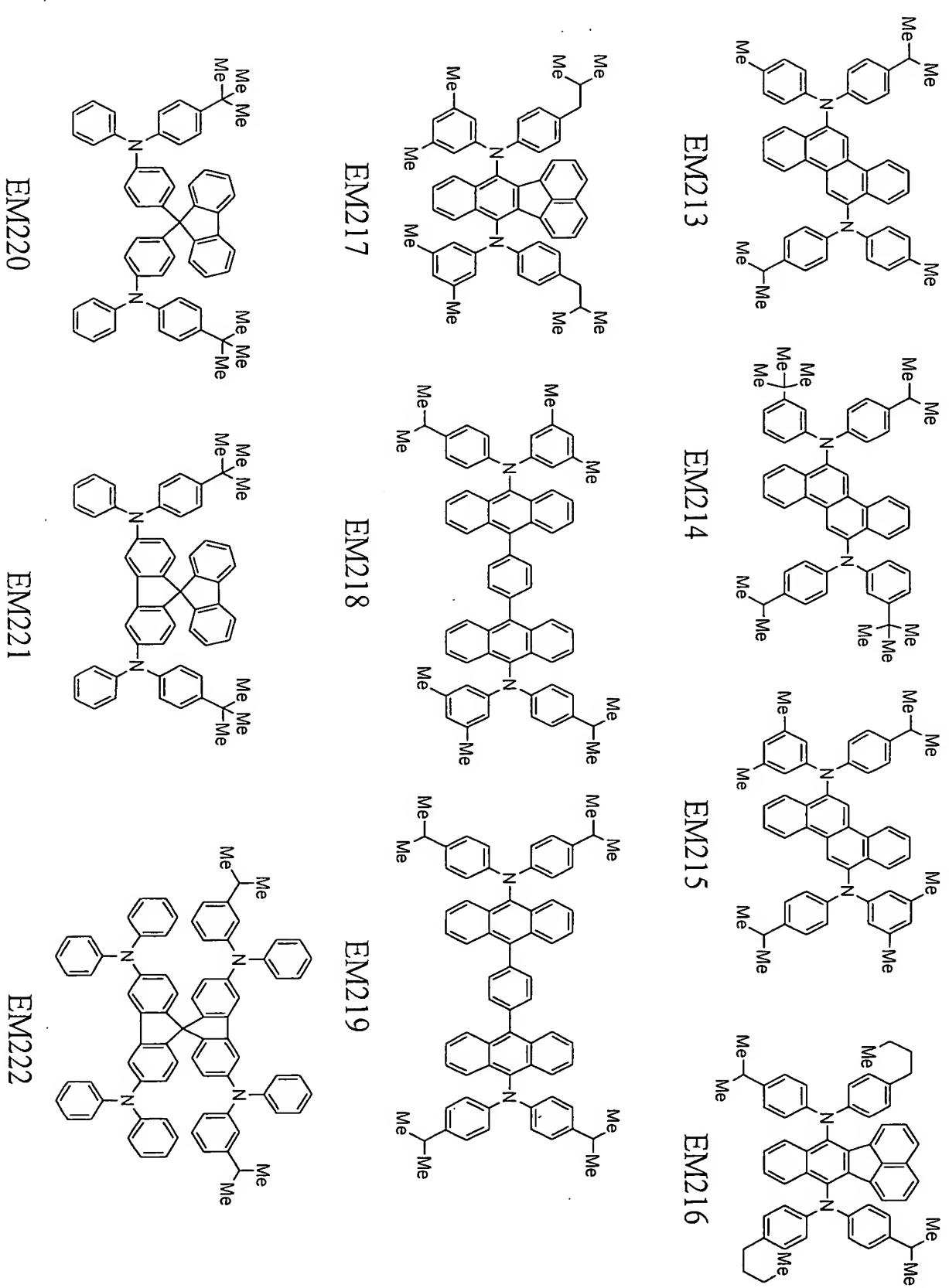


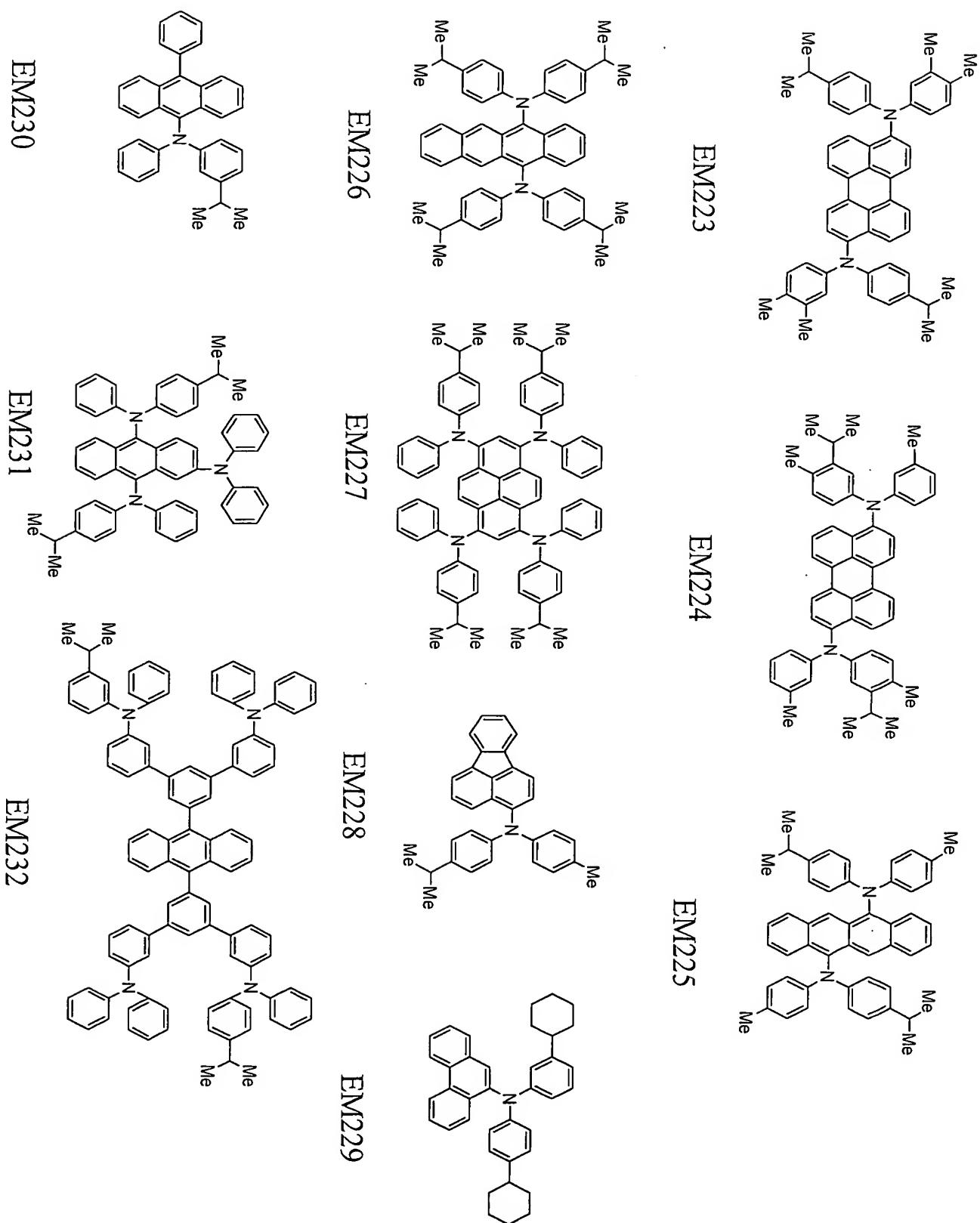
EM193



EM194







In the present invention, the ratio of the amount by weight of the arylamine of component (A) to the amount by weight of the anthracene derivative of component (B) in the layer of an organic light emitting medium is in the range of 1:99 to 99:1. It is preferable that the ratio of the amounts is suitably selected in accordance with the type of the compounds used. It is more preferable that, taking it into consideration that the compound of component (A) has the hole transporting property and the compound of component (B) has the electron transportation property, the ratio of the amounts is selected in a manner such that the life and the efficiency of the obtained device are maximized.

It is preferable that the ratio of the amount by weight of component (A) to the amount by weight of component (B) is in the range of 1:99 to 20:80. A particularly high efficiency can be obtained in this range.

It is preferable that the layer of an organic light emitting medium has a thickness in the range of 5 to 200 nm and more preferably in the range of 10 to 40 nm since the voltage applied to the device can be lowered to a great extent.

Due to the use of component (A) in combination with component (A) for the layer of an organic light emitting medium, the efficiency can be increased by 3 to 5 times as much as the efficiency obtained by using component (A) alone and the life can be increased by at least 3 times, and by at least 10 times when optimized, as long as the life obtained by using component (A) alone.

Due to the use of the arylamine represented by general formula (V) as component (A), the concentration quenching due to an increase in the association of molecules can be prevented since the steric hindrance

increases and the life can be further increased. When a branched alkyl group is introduced into the substituent of amino group or the condensed aromatic ring, the half width of the spectrum of the emitted light which can be used as the index for the purity of color is decreased since the steric 5 repulsion between the condensed aromatic ring and the substituent of amino group is increased and the spectrum of the emitted light becomes sharp. Therefore, the obtained device is suitable for full color displays.

Due to the use of component (A) and component (B) in combination, the stability and the heat resistance are improved since the layer of an 10 organic light emitting layer becomes more amorphous. It is preferable that the compound of component (B) has a glass transition temperature of 110°C or higher. It is also preferable that the compound of component (A) has a glass transition temperature of 70°C or higher. By mixing the compounds having the above glass transition temperatures, the glass 15 transition temperature of the layer of an organic light emitting medium can be made 90°C or higher and a durability in storage of 500 hours or longer at 85°C can be achieved.

The organic EL device of the present invention comprises the layer 20 of an organic light emitting medium (referred to as the light emitting medium layer, hereinafter) which comprises the combination of component (A) and component (B) and is disposed between a pair of electrodes. In the organic EL device of the present invention, it is preferable that various intermediate layers are disposed between the electrodes and the light emitting medium layer. Examples of the intermediate layer include a 25 hole injecting layer, a hole transporting layer, an electron injecting layer and an electron transporting layer. It is known that various organic and

inorganic compounds can be used for these layers.

Typical examples of the construction of the organic EL device include:

- (1) An anode / a light emitting layer / a cathode;
- 5 (2) An anode / a hole injecting layer / a light emitting layer / a cathode;
- (3) An anode / a light emitting layer / an electron injecting layer / a cathode;
- (4) An anode / a hole injecting layer / a light emitting layer / an electron injecting layer / a cathode;
- 10 (5) An anode / an organic semiconductor layer / a light emitting layer / a cathode;
- (6) An anode / an organic semiconductor layer / an electron barrier layer / a light emitting layer / a cathode;
- (7) An anode / an organic semiconductor layer / a light emitting layer / an adhesion improving layer / a cathode;
- 15 (8) An anode / a hole injecting layer / a hole transporting layer / a light emitting layer / an electron injecting layer / a cathode;
- (9) An anode / an insulating layer / a light emitting layer / an insulating layer / a cathode;
- 20 (10) An anode / an inorganic semiconductor layer / an insulating layer / a light emitting layer / an insulating layer / a cathode;
- (11) An anode / an organic semiconductor layer / an insulating layer / a light emitting layer / an insulating layer / a cathode;
- (12) An anode / an insulating layer / a hole injecting layer / a hole transporting layer / a light emitting layer / an insulating layer / a cathode;
- 25 and

(13) An anode / an insulating layer / a hole injecting layer / a hole transporting layer / a light emitting layer / an electron injecting layer / a cathode.

Among the above constructions, construction (8) is preferable.  
5 However, the construction of the organic EL device is not limited to the above examples.

In general, the organic EL device is prepared on a substrate which transmits light. The substrate which transmits light is the substrate which supports the organic EL device. It is preferable that the substrate 10 which transmits light has a transmittance of light of 50% or greater in the visible region of 400 to 700 nm. It is also preferable that a flat and smooth substrate is used.

As the substrate which transmits light, for example, glass plates and synthetic resin plates are advantageously used. Specific examples of 15 the glass plate include plates made of soda ash glass, glass containing barium and strontium, lead glass, aluminosilicate glass, borosilicate glass, barium borosilicate glass and quartz. Specific examples of the synthetic resin plates include plates made of polycarbonate resins, acrylic resins, polyethylene terephthalate resins, polyether sulfide resins and polysulfone 20 resins.

As the anode, an electrode made of a material such as a metal, an alloy, a conductive compound and a mixture of these materials which has a great work function (4 eV or more) is preferably used. Specific examples of the material for the anode include metals such as Au and conductive 25 materials such as CuI, ITO (indium tin oxide), SnO<sub>2</sub>, ZnO and In-Zn-O. The anode can be prepared by forming a thin film of the electrode material

described above in accordance with a process such as the vapor deposition process and the sputtering process. When the light emitted from the light emitting layer is obtained through the anode, it is preferable that the anode has a transmittance of the emitted light greater than 10%. It is 5 also preferable that the sheet resistivity of the anode is several hundred  $\Omega/\square$  or smaller. The thickness of the anode is, in general, selected in the range of 10 nm to 1  $\mu\text{m}$  and preferably in the range of 10 to 200 nm although the preferable range may be different depending on the used material.

10 As the cathode, an electrode made of a material such as a metal, an alloy, a conductive compound and a mixture of these materials which has a small work function (4 eV or smaller) is used. Specific examples of the material for the cathode include sodium, sodium-potassium alloys, magnesium, lithium, magnesium-silver alloys, aluminum/aluminum 15 oxide,  $\text{Al/Li}_2\text{O}$ ,  $\text{Al/LiO}_2$ ,  $\text{Al/LiF}$ , aluminum-lithium alloys, indium and rare earth metals.

The cathode can be prepared by forming a thin film of the electrode material described above in accordance with a process such as the vapor deposition process and the sputtering process.

20 When the light emitted from the light emitting medium layer is obtained through the cathode, it is preferable that the cathode has a transmittance of the emitted light greater than 10 %. It is also preferable that the sheet resistivity of the cathode is several hundred  $\Omega/\square$  or smaller. The thickness of the cathode is, in general, selected in the range of 10 nm 25 to 1  $\mu\text{m}$  and preferably in the range of 50 to 200 nm although the preferable range may be different depending on the material used.

In the organic EL device of the present invention, it is preferable that a layer of a chalcogenide, a metal halide or a metal oxide (this layer may occasionally be referred to as a surface layer) is disposed on the surface of at least one of the pair of electrodes prepared as described above.

5 Specifically, it is preferable that a layer of a chalcogenide (including an oxide) of a metal such as silicon and aluminum is disposed on the surface of the anode at the side of the light emitting medium layer and a layer of a metal halide or a metal oxide is disposed on the surface of the cathode at the side of the light emitting medium layer. Due to the above layers,

10 stability in driving can be improved.

Preferable examples of the chalcogenide include  $\text{SiO}_x$  ( $1 \leq x \leq 2$ ),  $\text{AlO}_x$  ( $1 \leq x \leq 1.5$ ),  $\text{SiON}$  and  $\text{SiAlON}$ . Preferable examples of the metal halide include  $\text{LiF}$ ,  $\text{MgF}_2$ ,  $\text{CaF}_2$  and fluorides of rare earth metals. Preferable examples of the metal oxide include  $\text{Cs}_2\text{O}$ ,  $\text{Li}_2\text{O}$ ,  $\text{MgO}$ ,  $\text{SrO}$ ,

15  $\text{BaO}$  and  $\text{CaO}$ .

In the organic EL device of the present invention, the electron transporting property and the hole transporting property of the light emitting medium layer are simultaneously improved by suitably adjusting the relative amounts of component (A) and component (B) described above and the above intermediate layers such as the hole injecting layer, the hole transporting layer and the electron injecting layer can be omitted. In this case, it is preferable that the surface layer described above is disposed.

In the organic EL device of the present invention, it is preferable that a mixed region of an electron transfer compound and a reducing dopant or a mixed region of a hole transfer compound and an oxidizing dopant is disposed on the surface of at least one of the pair of electrodes

prepared as described above. Due to the mixed region disposed as described above, the electron transfer compound is reduced to form an anion and injection and transportation of electrons from the mixed region into the light emitting medium can be facilitated. The hole transfer 5 compound is oxidized to form a cation and injection and transportation of holes from the mixed region into the light emitting medium is facilitated. Preferable examples of the oxidizing dopant include various types of Lewis acid and acceptor compounds. Preferable examples of the reducing dopant include alkali metals, compounds of alkali metals, alkaline earth 10 metals, rare earth metals and compounds of these metals.

In the organic EL device of the present invention, the light emitting medium layer has the following functions:

- (1) The injecting function: the function of injecting holes from the anode or the hole injecting layer and injecting electrons from the cathode or the 15 electron injecting layer when an electric field is applied;
- (2) The transporting function: the function of transporting injected charges (electrons and holes) by the force of the electric field; and
- (3) The light emitting function: the function of providing the field for recombination of electrons and holes and leading the recombination to the 20 emission of light.

As the process for forming the light emitting medium layer, a conventional process such as the vapor deposition process, the spin coating process and the Langmuir-Blodgett process (the LB process) can be used. It is particularly preferable that the organic light emitting medium layer is 25 a molecular deposit film. The molecular deposit film is a thin film formed by deposition of a material compound in the gas phase or a thin film

formed by solidification of a material compound in a solution or in the liquid phase. In general, the molecular deposit film can be distinguished from the thin film formed in accordance with the LB process (the molecular accumulation film) based on the differences in the aggregation structure and higher order structures and functional differences caused by these structural differences.

As disclosed in Japanese Patent Application Laid-Open No. Showa 57(1982)-51781, the light emitting medium layer can also be formed by dissolving a binder such as a resin and the material compounds into a solvent to prepare a solution, followed by forming a thin film from the prepared solution in accordance with the spin coating process or the like.

In the present invention, where desired, the light emitting medium layer may comprise conventional organic light emitting media other than component (A) and component (B) described above or the light emitting medium layer comprising the compounds described in the present invention may be laminated with a light emitting medium layer comprising other conventional organic light emitting media as long as the object of the present invention is not adversely affected.

The hole injecting layer and the hole transporting layer are layers which help injection of holes into the light emitting medium layer and transport the holes to the light emitting region. The layers exhibit a great mobility of holes and, in general, have an ionization energy as small as 5.5 eV or smaller. For the hole injecting layer and the hole transporting layer, a material which transports holes to the light emitting medium layer at a small electric field strength is preferable. A material which exhibits, for example, a mobility of holes of at least  $10^{-6} \text{ cm}^2/\text{V}\cdot\text{sec}$  under application of

an electric field of  $10^4$  to  $10^6$  V/cm is more preferable. A material can be selected from materials which are conventionally used as the charge transporting material of holes in photoconductive materials and conventional materials which are used for the hole injecting layer in  
5 organic EL devices.

To form the hole injecting layer or the hole transporting layer, a thin film may be formed from a material for the hole injecting layer or the hole transporting layer, respectively, in accordance with a conventional process such as the vacuum vapor deposition process, the spin coating process, the  
10 casting process and the LB process. The thickness of the hole injecting layer and the hole transporting layer is not particularly limited. In general, the thickness is 5 nm to 5  $\mu$ m.

The electron injection layer is a layer which helps injection of electrons into the light emitting medium layer and exhibits a great  
15 mobility of electrons. The adhesion improving layer is a layer made of a material exhibiting excellent adhesion with the cathode among the electron injecting layer. As the material for the electron injecting layer, metal complexes of 8-hydroxyquinoline and derivatives thereof are preferably used. Specific examples of the metal complex of 8-hydroxyquinoline and derivatives thereof include metal chelates of oxinoid  
20 compounds including chelates of oxine (in general, 8-quinolinol or 8-hydroxyquinoline). For example, tris(8-quinolinol)aluminum can be used as the electron injecting material.

The organic EL device of the present invention tends to form defects  
25 in pixels due to leak and short circuit since an electric field is applied to ultra-thin films. To prevent the formation of the defects, a layer of an

insulating thin film may be inserted between the pair of electrodes.

Examples of the material used for the insulating layer include aluminum oxide, lithium fluoride, lithium oxide, cesium fluoride, cesium oxide, magnesium oxide, magnesium fluoride, calcium oxide, calcium fluoride, aluminum nitride, titanium oxide, silicon oxide, germanium oxide, silicon nitride, boron nitride, molybdenum oxide, ruthenium oxide and vanadium oxide. Mixtures and laminates of the above compounds can also be used.

To prepare the organic EL device of the present invention, for example, the anode, the light emitting medium layer and, where necessary, the hole injecting layer and the electron injecting layer are formed in accordance with the above process using the above materials and the cathode is formed in the last step. The organic EL device may be prepared by forming the above layers in the order reverse to that described above, i.e., the cathode being formed in the first step and the anode in the last step.

An embodiment of the process for preparing an organic EL device having a construction in which an anode, a hole injecting layer, a light emitting medium layer, an electron injecting layer and a cathode are disposed successively on a substrate which transmits light will be described in the following.

On a suitable substrate which transmits light, a thin film made of a material for the anode is formed in accordance with the vapor deposition process or the sputtering process so that the thickness of the formed thin film is 1  $\mu\text{m}$  or smaller and preferably in the range of 10 to 200 nm. The formed thin film is used as the anode. Then, a hole injecting layer is

formed on the anode. The hole injecting layer can be formed in accordance with the vacuum vapor deposition process, the spin coating process, the casting process or the LB process, as described above. The vacuum vapor deposition process is preferable because a uniform film can  
5 be easily obtained and the possibility of formation of pin holes is small. When the hole injecting layer is formed in accordance with the vacuum vapor deposition process, in general, it is preferable that the conditions are suitably selected in the following ranges: the temperature of the source of the deposition: 50 to 450°C; the vacuum:  $10^{-7}$  to  $10^{-3}$  Torr; the rate of  
10 deposition: 0.01 to 50 nm/second; the temperature of the substrate: -50 to 300°C and the thickness of the film: 5 nm to 5  $\mu$ m; although the conditions of the vacuum vapor deposition are different depending on the used compound (the material for the hole injecting layer) and the crystal structure and the recombination structure of the hole injecting layer to be  
15 formed.

Then, the light emitting medium layer is formed on the hole injecting layer formed above. Using the organic light emitting medium described in the present invention, a thin film of the organic light emitting medium can be formed in accordance with the vacuum vapor deposition process, the sputtering process, the spin coating process or the casting process and the formed thin film is used as the light emitting medium layer. The vacuum vapor deposition process is preferable because a uniform film can be easily obtained and the possibility of formation of pin holes is small. When the light emitting medium layer is formed in accordance with the vacuum vapor deposition process, in general, the conditions of the vacuum vapor deposition process can be selected in the  
20  
25

same ranges as those described for the vacuum vapor deposition of the hole injecting layer although the conditions are different depending on the used compound. It is preferable that the thickness is in the range of 10 to 40 nm.

5 An electron injecting layer is formed on the light emitting medium layer formed above. Similarly to the hole injecting layer and the light emitting medium layer, it is preferable that the electron injecting layer is formed in accordance with the vacuum vapor deposition process since a uniform film must be obtained. The conditions of the vacuum vapor 10 deposition can be selected in the same ranges as those described for the vacuum vapor deposition of the hole injecting layer and the light emitting medium layer.

A cathode is formed on the electron injecting layer formed above in the last step and an organic EL device can be obtained. The cathode is 15 made of a metal and can be formed in accordance with the vacuum vapor deposition process or the sputtering process. It is preferable that the vacuum vapor deposition process is used in order to prevent formation of damages on the lower organic layers during the formation of the film.

In the above preparation of the organic EL device, it is preferable 20 that the above layers from the anode to the cathode are formed successively while the preparation system is kept in a vacuum after being evacuated.

The organic EL device which can be prepared as described above 25 emits light when a direct voltage of 3 to 40 V is applied in the condition that the anode is connected to a positive electrode (+) and the cathode is connected to a negative electrode (-). When the connection is reversed, no

electric current is observed and no light is emitted at all. When an alternating voltage is applied to the organic EL device, light emission is observed only in the condition that the polarity of the anode is positive and the polarity of the cathode is negative. When an alternating voltage is 5 applied to the organic EL device, any type of wave shape can be used.

The present invention also provides the organic light emitting medium comprising component (A) and component (B) described above. The organic light emitting medium is advantageously used for the organic electroluminescence device having excellent heat resistance and a long life 10 and efficiently emitting bluish to yellowish light.

The present invention will be described more specifically with reference to examples in the following. However, the present invention is not limited to the examples.

### 15 Example 1

On a glass plate having a size of  $25 \times 75 \times 1.1$  mm, a transparent electrode made of indium tin oxide and having a thickness of 120 nm was formed. After the glass substrate was cleaned by irradiation with ultraviolet light and exposure to ozone, the glass substrate was placed in a 20 vacuum vapor deposition apparatus.

After N,N'-bis[4-(diphenylamino)phenyl]-N,N'-diphenylbiphenyl-4,4'-diamine was vapor deposited so that a hole injecting layer having a thickness of 60 nm was formed, N,N,N',N'-tetrakis(4-biphenyl)-4,4'-benzidine was vapor deposited on the formed hole injecting layer so that a 25 hole transporting layer having a thickness of 20 nm was formed. Then, compound (EM4) shown above as component (B) and compound (EM83)

shown above as component (A) were simultaneously vapor deposited in amounts such that the ratio of the amount by weight of component (B) to the amount by weight of component (A) was 40:3 so that a light emitting layer having a thickness of 40 nm was formed.

5 On the formed light emitting layer, tris(8-hydroxyquinolinato)-aluminum (Alq) was vapor deposited so that an electron injecting layer having a thickness of 20 nm was formed. Lithium fluoride (LiF) was vapor deposited so that a layer having a thickness of 0.3 nm was formed and then aluminum (Al) was vapor deposited so that a layer having a 10 thickness of 150 nm was formed. The formed LiF/Al film worked as the cathode. An organic EL device was prepared as described above.

15 The prepared organic EL device was tested by passing an electric current. Light emission of pure blue color (the half width: 42 nm) having a luminance of 205 cd/m<sup>2</sup> was obtained at a voltage of 6.5 V and a current density of 10 mA/cm<sup>2</sup>. The device was tested by continuously passing a direct current at an initial luminance of 500 cd/m<sup>2</sup> and the half-life time was found to be 900 hours.

#### Examples 2 to 19

20 Organic EL devices were prepared in accordance with the same procedures as those conducted in Example 1 except that compounds shown in Table 1 were used as component (B) and component (A). The results of testing the obtained devices by passing an electric current at a current density of 10 mA/cm<sup>2</sup> are shown in Table 1. The half-life times obtained 25 by the test of continuous passage of a direct current at initial luminances shown in Table 1 are also shown in Table 1.

### Comparative Example 1

An organic EL device was prepared in accordance with the same procedures as those conducted in Example 1 except that a light emitting 5 layer having a thickness of 40 nm was formed with compound (EM4) alone in place of the combination of compound (EM4) and compound (EM83) used in Example 1. The results of testing the obtained device by passing an electric current at a current density of 10 mA/cm<sup>2</sup> are shown in Table 1. The device was tested by continuously passing a direct current at an initial 10 luminance of 500 cd/m<sup>2</sup> and the half-life time was found to be as short as 90 hours.

### Comparative Example 2

An organic EL device was prepared in accordance with the same 15 procedures as those conducted in Example 1 except that 4,4'-bis(diphenylamino)stilbene (H2) was used in place of compound (EM83) used in Example 1. The results of testing the obtained device by passing an electric current at a current density of 10 mA/cm<sup>2</sup> are shown in Table 1. The device was tested by continuously passing a direct current at an initial 20 luminance of 500 cd/m<sup>2</sup> and the half-life time was found to be as short as 300 hours.

### Comparative Example 3

An organic EL device was prepared in accordance with the same 25 procedures as those conducted in Example 1 except that 2,5,8,11-tetra-t-butylperylene (H3) was used in place of compound (EM83) used in

Example 1. The results of testing the obtained device by passing an electric current at a current density of 10 mA/cm<sup>2</sup> are shown in Table 1. The device was tested by continuously passing a direct current at an initial luminance of 1,000 cd/m<sup>2</sup> and the half-life time was found to be as short as 5 200 hours.

#### Comparative Example 4

An organic EL device was prepared in accordance with the same procedures as those conducted in Example 1 except that N,N'-di(naphthalen-2-yl),N,N'-diphenylbenzene (H4) was used in place of compound (EM83) used in Example 1. The results of testing the obtained device by passing an electric current at a current density of 10 mA/cm<sup>2</sup> are shown in Table 1. The device was tested by continuously passing a direct current at an initial luminance of 500 cd/m<sup>2</sup> and the half-life time was 15 found to be as short as 200 hours.

#### Comparative Example 5

An organic EL device was prepared in accordance with the same procedures as those conducted in Example 11 except that 1,3-bis- [2-{4-20 N,N'-(diphenylamino)phenyl}vinyl]benzene (H5) was used in place of compound (EM98) used in Example 11. The results of testing the obtained device by passing an electric current at a current density of 10 mA/cm<sup>2</sup> are shown in Table 1. The device was tested by continuously passing a direct current at an initial luminance of 1,000 cd/m<sup>2</sup> and the 25 half-life time was found to be as short as 750 hours.

### Example 20

An organic EL device was prepared in accordance with the same procedures as those conducted in Example 1 except that compounds shown in Table 2 were used as component (B) and component (A) and Alq:Cs/Al 5 was used as the cathode. Alq:Cs/Al was a mixed layer containing Alq and Cs (cesium) metal as the electron transporting compounds in relative amounts by mole of 1:1. The results of testing the obtained device by passing an electric current at a current density of 10 mA/cm<sup>2</sup> are shown in Table 2. The half-life time obtained by the test of continuous passage of a 10 direct current at the initial luminance shown in Table 2 is also shown in Table 2.

### Examples 21 and 22

An organic EL device was prepared in accordance with the same 15 procedures as those conducted in Example 20 except that compounds shown in Table 2 were used as component (B) and component (A). The results of testing the obtained devices by passing an electric current at a current density of 10 mA/cm<sup>2</sup> are shown in Table 2. The half-life times obtained by the test of continuous passage of a direct current at initial 20 luminances shown in Table 2 are also shown in Table 2.

Table 1 - 1

5	Components of light emitting layer		Voltage (V)	Luminance of emitted light (cd/m <sup>2</sup> )	Efficiency of light emission (cd/A)	
	component (B)	component (A)				
10	Example 1	EM4	EM83	6.5	205	2.05
10	Example 2	EM4	EM110	6.3	310	3.10
10	Example 3	EM5	EM111	6.0	325	3.25
10	Example 4	EM5	EM77	6.8	195	1.95
10	Example 5	EM5	EM117	6.0	295	2.95
10	Example 6	EM27	EM110	6.5	190	1.90
15	Example 7	EM37	EM111	6.0	180	1.80
15	Example 8	EM43	EM110	6.2	165	1.65
15	Example 9	EM49	EM111	6.3	170	1.70
15	Example 10	EM4	EM60	6.0	350	3.50
15	Example 11	EM4	EM98	6.0	730	7.30
20	Example 12	EM5	EM60	6.0	345	3.45
20	Example 13	EM5	EM98	6.0	815	8.15
20	Example 14	EM5	EM97	6.5	5	5.50
20	Example 15	EM27	EM98	6.0	355	3.55
20	Example 16	EM42	EM97	6.5	395	3.95
25	Example 17	EM46	EM98	6.0	500	5.00
25	Example 18	EM4	EM89	7.0	1050	10.50
25	Example 19	EM4	EM94	7.5	950	9.50
30	Comparative Example 1	EM4		6.3	90	0.90
30	Comparative Example 2	EM4	H2	6.8	105	1.05
30	Comparative Example 3	EM4	H3	6.5	200	2.00
35	Comparative Example 4	EM4	H4	7.0	96	0.96
35	Comparative Example 5	EM4	H5	6.8	510	5.10

Table 1 - 2

	Color of emitted light	Half width (nm)	Half-life time (hour)	Initial luminance (cd/m <sup>2</sup> )
5				
	Example 1	pure blue	42	900
	Example 2	pure blue	43	2900
10	Example 3	pure blue	45	3050
	Example 4	pure blue	42	680
	Example 5	pure blue	44	1000
	Example 6	pure blue	44	1150
	Example 7	pure blue	45	1200
15	Example 8	pure blue	43	950
	Example 9	pure blue	45	1100
	Example 10	blue	40	800
	Example 11	blue	50	3100
	Example 12	blue	41	700
20	Example 13	blue	49	3200
	Example 14	blue	48	2950
	Example 15	blue	49	1500
	Example 16	blue	49	900
	Example 17	blue	50	1000
25	Example 18	green	68	1050
	Example 19	green	65	750
	Comparative Example 1	pure blue	45	90
30	Comparative Example 2	pure blue	58	300
	Comparative Example 3	green	69	200
35	Comparative Example 4	pure blue	46	100
	Comparative Example 5	blue	62	500
				1000

Table 2 - 1

5	Components of light emitting layer		Voltage (V)	Luminance of emitted light (cd/m <sup>2</sup> )	Efficiency of light emission (cd/A)
	component (B)	component (A)			
10	Example 20	EM32	EM111	5.0	290
	Example 21	EM4	EM128	6.5	260
	Example 22	EM5	EM128	6.0	250
	Example 23	EM42	EM128	6.5	155
	Example 24	EM5	EM131	6.7	964
15	Example 25	EM5	EM133	6.5	1015
	Example 26	EM43	EM139	7.0	950
	Example 27	EM5	EM139	6.5	2040
	Example 28	EM42	EM144	6.5	1050
	Example 29	EM5	EM144	6.4	2100
20	Example 30	EM32	EM144	6.0	1555
	Example 31	EM32	EM160	6.5	1430
	Example 32	EM32	EM189	6.0	980
	Example 33	Alq	EM139	7.0	1420
	Example 34	EM4	EM215	6.5	340
25	Example 35	EM5	EM215	6.5	355
	Example 36	EM42	EM215	6.7	185
	Example 37	EM4	EM195	6.0	1050
	Example 38	EM4	EM197	6.4	1030
	Example 39	EM5	EM202	6.5	1870
30	Example 40	EM5	EM204	6.5	1850
	Example 41	EM5	EM208	6.9	1350

Table 2 - 2

		Color of emitted light	Half width (nm)	Half-life time (hour)	Initial luminance (cd/m <sup>2</sup> )
5					
	Example 20	pure blue	44	1300	500
	Example 21	pure blue	43	2100	500
	Example 22	pure blue	43	2350	500
10	Example 23	pure blue	44	1000	500
	Example 24	blue	49	4000	1000
	Example 25	blue	50	4100	1000
	Example 26	green	68	900	3000
	Example 27	green	67	4500	3000
15	Example 28	green	67	1100	3000
	Example 29	green	68	4750	3000
	Example 30	green	68	1050	3000
	Example 31	green	64	1800	3000
	Example 32	green	65	950	3000
20	Example 33	green	69	1500	3000
	Example 34	pure blue	44	3500	500
	Example 35	pure blue	44	3950	500
	Example 36	pure blue	43	970	500
	Example 37	blue	50	4050	1000
25	Example 38	blue	49	3950	1000
	Example 39	green	68	2100	3000
	Example 40	green	68	1950	3000
	Example 41	green	65	1500	3000

30

As shown in Table 1 and Table 2, the excellent efficiencies and the excellent lives could be achieved in the devices emitting green light, blue light and pure blue light, which was hard to obtain, as shown in Examples 35 1 to 41. The above results were achieved since the light having more

excellent purity of color could be emitted due to the decrease in the half width in comparison to those of the devices of Comparative Examples.

In particular, the devices comprising the diaminoanthracene derivatives emitting green light, the diaminopyrene derivatives emitting 5 blue light and the diaminochrysene derivatives emitting pure blue light as component (A) exhibited more excellent efficiencies of light emission and lives in comparison to those of any of the devices of Comparative Examples.

Since the anthracene derivative was used as component (B) and the 10 diaminoanthracene derivative, the diaminopyrene derivative or the diaminochrysene derivative was used as component (A), the most excellent efficiency of light emission and life could be achieved by the devices emitting green light, blue light and pure blue light.

## 15 INDUSTRIAL APPLICABILITY

In accordance with the present invention, the organic EL device which exhibits excellent purity of color, has excellent heat resistance and a long life and efficiently emits bluish to yellowish light and an organic light emitting medium which can be advantageously used for the organic 20 electroluminescence device, can be provided.

The organic EL device can be advantageously used as the light emitting device in various types of display apparatuses and is particularly suitable for full color display apparatuses.